THE PROCEEDINGS OF THE PHYSICAL SOCIETY

Section A

VOL. 63, PART 4 1 April 1950 No. 364 A

Rutherford Memorial Lecture (1948)

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4th Rutherford Memorial Lecture, delivered 14th December 1949;
MS. received 19th December 1949

LORD RUTHERFORD died in 1937, 12 years ago, and even at that date it was sufficiently clear that from the perspective of world history his life's work constituted one of the upsurges of knowledge and of the understanding of natural forces, comparable in importance to those inaugurated by Newton and by Faraday. Apart, however, from the applications in modern 'electronics' of the basic discoveries underlying the ionization theory of the passage of electricity through gases which he shared with J. J. Thomson and others, it was at that time hardly possible to forecast that Rutherford's own particular work on radioactive transformations and radiations would be applied and exploited so speedily and in so spectacular a manner as occurred at Hiroshima in 1945, or, for example, that the important uses of radioactive isotopes would have developed to the extent to which they are used even at present.

It may perhaps be desirable to qualify this latter statement somewhat, since Rutherford himself, though cautious by nature and not in the habit of making claims greater than warranted by the experimental evidence immediately available, stated somewhat prophetically in the Watt Anniversary Lecture in January 1936, in discussing the possibility of obtaining energy from atomic transformations: "The recent discovery of the neutron and the proof of its extraordinary effectiveness in producing transformations (of atoms) at very low velocities opens up new possibilities, if only a method could be found of producing slow neutrons in quantity with little expenditure of energy." It is interesting to note that it was this latter development which later actually formed the basis for the application of atomic energy.

It is now more evident that his work has introduced a new scientific era, both in the fundamental scientific concepts involved, and in the pioneering of atomic energy. Moreover, the social consequences are likely to be such that future historians *, looking back on this period, will assuredly be interested in the environment which produced such a man, and the conditions which enabled such advances to be made so largely as the result of the work and inspiration of that one man. It seems appropriate, therefore, that we should endeavour to hand down a true account to posterity. In Rutherford's own words in 1936 and applied to others preceding him, "We all ought to give some of our recollections of those past and gone, before they are lost for good."* Such, I take it, is the special duty of those called upon to give the earlier of this series of Rutherford Memorial Lectures, and I propose in this regard to follow the example of my predecessors.

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* G. M. Trevelyan, quoted in *The Times* of 1st December 1949 : " There is too little about science quoted in our histories, considering that science has been the chief factor in human affairs, and particularly English affairs, during the past 200 years ".

In the time at our disposal this evening, however, it is not possible for me to traverse recollections or information gleaned relating to the whole period of his life and vast extent of his work. I propose, therefore, to confine myself in the main to two special periods of his life with which it was my good fortune to come more especially into contact: firstly, his early life and training in New Zealand, and secondly, the circumstances and experiments surrounding the evolution of the idea of the nuclear atom, and of the first experiments in which the disintegration of nitrogen was observed or deduced. As you are no doubt well aware the latter was the starting point in the studies of artificial transmutation of atoms.

It may be, that in devoting a large proportion of this talk to his early life I should ask your indulgence because I shall need to go back 100 years. I have come to the conviction, however, that in comparing Rutherford's work with that of his great scientific predecessors it will be found that, added to the application of his high degree of natural genius, its success lay in his personal human characteristics of natural exuberant energy and enthusiasm, in his capacity to see an objective and proceed to it in the most direct method with the materials at hand, and in his selflessness and force of character in his relation to colleagues—qualities all inculcated in his unique early home and environment—these were the main factors in his success and in his ability to gather round him willing co-operators, working and serving with loyalty and affection. Thus he was able to cause a spirit of enquiry to glow in every breast, every new discovery led to a further investigation by one or other, and each member of the lucky teams in succession at Montreal, Manchester and Cambridge became zealous to distinguish himself in the common pursuit.

The story of Rutherford's early life and training is interwoven with the epic story of the early colonization of New Zealand: a story as fascinating and romantic as that of any period of Britain's long history.

Ernest Rutherford was born in 1871, but to understand his early environment we need to go back to that of his parents, for it was to the parents' influence and particularly the mother that we owe most of Rutherford's success, as he himself testified on very many occasions, as witness, for example, the cable he sent to his old mother in 1931 on his elevation to the peerage—"Now Lord Rutherford. Honour more yours than mine. Love, Ernest".

It is perhaps difficult for a London city audience at this date to visualize the primitive natural conditions in the Nelson district at the time of Rutherford's birth, and in an attempt to convey the picture it is perhaps desirable briefly to recount the story of the settlement of that district in 1842—the year in which his father arrived. It is not generally realized that Nelson, and indeed most of the main provinces of New Zealand, were colonized by private enterprise organizations. Fortunately, in all of these, thanks to Wakefield, there was much practical idealism, albeit accompanied in the prospectus by rashly optimistic statements—probably not deliberately so, but tempting to would-be settlers. Nelson was the third of the colonization ventures of the New Zealand Company, and in its establishment it benefited from the examples of the earlier one at Wellington and its off-shoot

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* Quoted by N. Feather, *Lord Rutherford* (London: Blackie), p. 57.

New Plymouth, which in turn had gained from the experiences of colonization in Australia and from Lord Durham's report on Canadian settlements.

I need not dwell on the still earlier history. Tasman first saw and gave the name to New Zealand in 1642, and the first place at which he anchored and tried to land was in Golden Bay or Murderers Bay as he called it, in the Nelson Province, where four of his men were killed by the Maoris. This gave him such a respect for the fighting qualities of the natives that he did not make any further serious attempts to land there or elsewhere in New Zealand. At that time there was a large Maori population. Next came Captain James Cook in 1769 who, accompanied by the scientists Joseph Banks and Solander, rediscovered New Zealand. He charted the main coastline and made landings at several places. Later came missionaries in the North, and whalers who established depots around the Cook Straits. Gradually certain of the Maori tribes secured muskets, mostly bartered at the rate of one ton of dressed flax (*phormium tenax*) per musket, and great slaughter followed in the inter-tribal forays, particularly those of Te Rauparaha. This, together with the introduction of influenza decimated the population of the Cook Straits area, and the general feeling of insecurity made the remaining Maoris all the more ready to sell their ancestral land claims for a few paltry articles of trade to the precursors of the agents of the New Zealand Company, just before the latter arrived to fix the site of the Nelson settlement.

It is interesting to remember that the Colony of Nelson was planned in England, and the emigrant ships despatched without the site having been located even to within 100 miles. The settlement was intended to consist of 1,100 sections of 200 acres each, at a price of thirty shillings per acre. The priority of land was to be determined by ballot on arrival. The money raised by the sale of land was to be used as follows: £150,000 for conveying emigrants to the Colony, £100,000 for expenses and remuneration of the Company in selecting the site and otherwise promoting the safe and orderly establishment of the Colony, £15,000 towards religious endowments, £15,000 for founding a College, £20,000 for encouragement to steam navigation.

Thus the first three small sailing ships containing emigrants arrived in Wellington Harbour and after arguments with the British Governor, Hobson, who wished them to settle in the North, their advance party proceeded to the area to be called Nelson, whose coastline had been described and roughly mapped by the French explorer d'Urville some 14 years earlier in 1827. There a good harbour was discovered behind a 'boulder bank' almost by accident in October 1841, and shortly afterwards, in 1842, the main body arrived, disembarked and began to carve out homes in what fortunately proved to be beautiful land, though of limited area, and with a pleasant climate (Nelson boasts some 2,520 hours of sunshine per year, and yet with 40 inches of well distributed rainfall).

Thus in that year, April 1842, there arrived Rutherford's grandfather from Dundee, in the *Phoebe Dunbar* which sailed from Perth, with his father, James, then aged 3½, the former having been engaged by a Colonel Thoms to construct a sawmill etc.

The emigrants were of good type, attracted by the promising prospectus, and selected from sober, intelligent and mainly God-fearing applicants, most of them healthily dissatisfied with social conditions and opportunities in certain parts of the Old Country. They were knit together into a closely integrated community by the exigencies of the long perilous voyage (the *Phoebe Dunbar* had taken over

6½ months) and by the necessity for cooperation in facing the problems of settlement in the new land. Their only trouble with the Maoris was an unfortunate massacre of some 20 of them a year later at Wairau, due to misunderstanding as to the terms of purchase of some of the land taken up.

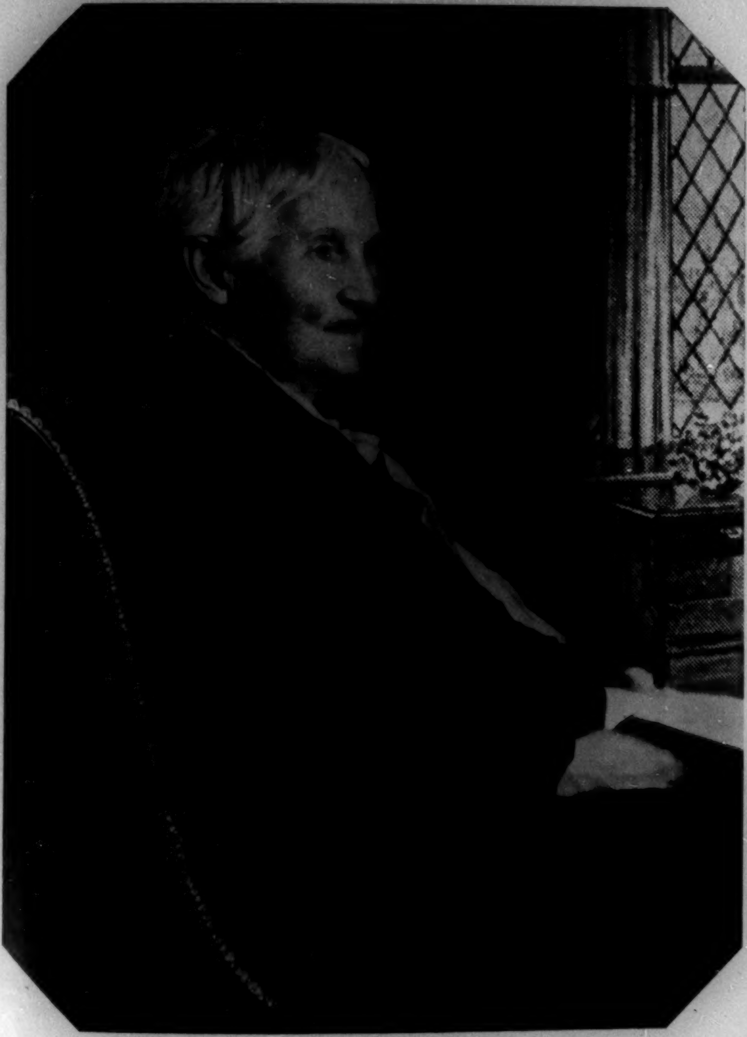
The ships in which these early settlers made the voyage measured perhaps 125 feet in length with a beam of 30 feet, and each carried some 200 emigrants, in addition to a crew of 30. The congestion can be imagined. Is it any wonder there was a high death rate, particularly amongst the children? Nevertheless, the white population of the town and district rose to 2,000 within the first year.

The Colonists had been selected as a cross section of the community from which they originated, large farmers, smallholders, labourers, artisans and members of the professions, but the area on which they settled was at first hemmed in by almost impassable mountains and they soon became more of a community of relatively small holders, though on the outskirts there were a few large holdings of poorer, hilly land used for sheep grazing. In these conditions there grew up a vigorous, healthy democracy in which the common good was the objective, which encouraged initiative and respected leadership, and in which each helped the other as a first principle of duty and loyalty, yet with a strong core of practical common sense rather than sentimentality or artificiality. Those of us who in later years were privileged to know Lord Rutherford will appreciate how much his social and political outlook reflected the influence of this early environment.

In spite of these primitive conditions the degree of mutual self-education and the provision of schools for children was pursued to a degree which is well-nigh astonishing. Only one year after the first landing, the local agent (John Ward) of the New Zealand Company, in his report on the progress of the settlement wrote asking for more school teachers, and could quote Dr. Canning as expressing the feelings of the Colonists as follows: "What we want is a body of teachers acquainted with the philosophy of the mind—gifted men and women who shall respect human nature in the child, and strive to teach and gently bring out his best powers and sympathies..... This requires that education shall be recognized by the community as its highest interest and duty. It requires that the instructors of youth should take the precedence of the money-getting classes. It requires that the parents shall sacrifice show and pleasure to the acquisition of the best possible helps and guides for their children."

I include this quotation because as we shall see, Rutherford's mother, who provided home teaching in school subjects, and the three school teachers (Ladley, Reynolds and Littlejohn in succession) who moulded his training, could fairly be described as fulfilling these conditions.

Lord Rutherford's father, James, grew to manhood in this primitive environment, while the land, formerly heavily bushed, was brought into agricultural production. Times were hard because there was little which could be produced for export until the Australian gold rush some 10 years later produced a demand for foodstuffs to be exported to that country. James Rutherford helped his father as a 'bush engineer', learning to improvise and not to be afraid of any job from that of wheelwright to bridge builder, using the native timbers and facilities immediately available. He had little school education, but devoured the popular magazines on mechanics as they arrived from U.K.; he learned to be a self-reliant, resourceful engineer even to the extent of developing water power, bravely and progressively practical in proceeding directly to the objectives in hand.



MRS. MARTHA RUTHERFORD.
AGED 90.

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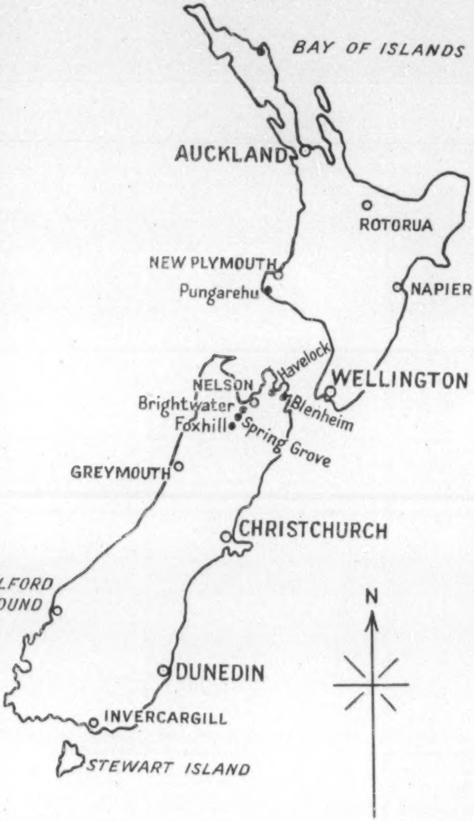
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Spring Grove
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In the meantime, in 1855, there had arrived in New Plymouth across the Cook Straits some 150 miles north, a youngish widow named Caroline Thompson (from Hornchurch, Essex) with her daughter Martha, aged 13, and her parents, who set up a windmill in that town. A few years later the Maori wars broke out in the North Island and women and children were evacuated in 1860 to the more peaceful Nelson district, and in succession Caroline and Martha became the first provincial women primary teachers at Spring Grove, near to where the Rutherfords had settled.

In this way James Rutherford and Martha Thompson met, and were married in 1866, having built themselves a house at Brightwater, some 12 miles south-west of Nelson town.

In this house they lived for 10 years, James Rutherford working as a small-scale farmer, wheelwright, engineer and 'flax'-miller, riding to his various jobs across the country tracks on a wooden bicycle of his own design and manufacture. There Ernest Rutherford was born in 1871, the fourth of a family of twelve.

James Rutherford appears to have resisted the lure of the various 'gold rushes' arising from discoveries in the back country around him and on the West Coast of the South Island. The family moved to a neighbouring hamlet at Foxhill, however, when Ernest was 5 years old, and there he attended the primary school until he reached the sixth standard at the age of 11. Even at that tender age he possessed a copy (shown) of a *Primer of Physics* by Balfour Stewart published in 1880, with his name and the date, 31st July 1882, written on the cover. The teacher, Ladley, was a fine man, able and conscientious.

James Rutherford had in the meantime extended his flaxmilling activities to a place named Ropaka near Havelock, some 35 miles away, and no doubt this was a factor in the decision of the parents to move to Havelock, at the head of Pelorus Sound. The roads were such that their effects had to be taken by boat.

At Havelock James Rutherford quickly built up assets as a result of his own efforts of hard work and ingenuity, and again from the capacity, necessary for success in such circumstances, for going directly for his objectives using the material and facilities at hand. This attitude was unconsciously handed over to Ernest and was characteristic of all his later scientific work.

Apart from his small domestic farm holding, James Rutherford had two main occupations, alternating with the seasons and sometimes overlapping. Firstly he set up a small sawmill where his main production was sleepers at 2*s*. 8*d*. each for the railways under intermittent construction in the Christchurch district, whence they were carried by small sailing ships, and secondly the cutting and 'stripping' or decortication of the native plant *phormium tenax* or New Zealand flax, for which there was a good overseas market for the production of ropes etc. This plant grew in the nearby swamps, it had tough broad leaves some five to six feet in length and some 2½ inches in breadth, with a fibre content of some 12%. Considerable ingenuity and skill were needed to put together and operate the rough machinery for the extraction of the fibre and its subsequent 'finishing' processes. This flaxcutting and milling compelled the father to work at relatively remote places, so that he could return to his home only at week-ends. The training of the family was therefore sometimes left largely to the mother.

The letters sent down to his wife during the working week bore striking witness to the complete understanding and affection between the parents. Theirs was at no stage a divided house. Sunday was a day of complete manual rest, when the mother would regularly assemble the family for religious exercises, she playing the piano and the father the violin, and for general mutual education such as spelling bees and geography, which was accompanied by the use of maps to illustrate current events and the news of the outside world contained in the newspapers to hand. One can trace to this period and influence the hymn tunes engrained on his subconscious mind with which in later life Rutherford occasionally and unknowingly used to regale his co-workers, as has been related elsewhere by Andrade and Robinson. Thus, while Rutherford inherited from his forbears a rich physical and mental endowment he was even more fortunate in the qualities and habits of work acquired from the example of his parents and the atmosphere of his home. In a recent talk on the Third Programme of the B.B.C., Professor F. J. Schonell has indicated the considerable extent to which stability of family life and home conditions are linked with learning, or as he puts it, "calm feeling makes clear thinking". A further relevant extract from Professor Schonell's lecture is as follows: "Language development depends on the general level of English in the home. Research shows that children of the same age and about the same intelligence coming from homes of widely different cultural levels and opportunities may differ by as much as 3 years in vocabulary; so great is the force of environment."

Rutherford's father was a man of great character, of fine, quiet disposition, straight and honourable. I have learned from many friends who visited the family either at Havelock or later at Pungarehu, of the many unostentatious acts of kindness extended—kindnesses of a nature which most leave undone or despise. How many of us later were indebted similarly to Lord and Lady Rutherford?

Martha Rutherford was a truly remarkable woman, of good education and character, very musical, a good organizer, thrifty and hard-working, yet above all human, happy and sunny in her relations with others. She was the family treasurer, but in spite of the demands on her purse the family on one occasion raised over £10 towards the Indian Famine Fund.

Mrs. Rutherford had at the time a governess, Miss Buckeridge, to assist her with the younger children, and she reports that "Ernest never needed to study; having read a school book once he knew it. He was a lively boy and I loved to see him and the other children at their mother's knee at prayers". Nevertheless her brother reports a later occasion when in a train he caught Ernest reading a 'yellow back' (wild west story).

Ernest, along with his brothers and sisters, attended the Havelock primary school and again they were fortunate in the teacher, Reynolds, otherwise the break in school training would have been more serious in its consequences to his career. The parents were ambitious for the children and, apart from the mother's home teaching, for a consideration Reynolds gave them extra lessons in Latin for an hour before the normal commencement of school. It soon became evident that Ernest in particular showed considerable promise, and the time came when he could compete for the Marlborough Provincial Scholarship, giving £40 per annum for residential board, plus free education at the Nelson College Secondary School. This College was modelled on the public schools in England at the time and had been founded in 1856 and endowed partly from land reserves and partly from the money (even though less was available than anticipated) set aside in the original charter of the New Zealand Company.

I was fortunate enough to be present in 1925 when Rutherford made a tour of the district and attended an evening meeting and reception in his honour by old scholars in the small village. It was a boisterously happy and jolly occasion. There was present the retired district inspector of schools, a Mr. Lambert, and in reminiscent vein he told the story of the rivalry between the various country schools and how the Havelock school authorities were ambitious to have one of their pupils gain the coveted scholarship. The pupils living in the larger centre of Blenheim had the advantage, additional to that derived from the size and number of teachers in their schools, because candidates from country schools had to be brought in and housed in Blenheim in unaccustomed surroundings, so that they might not be expected to give of their best in the examination. The Havelock school committee accordingly requested that their candidates be examined in their own school. This was granted by the Board and the inspector was deputed to supervise the examination, in December 1886. Great local interest had arisen, and when the day of examination arrived (a hot summer's day) interested residents would present themselves at the open door of the two-room school and in an anxious whisper ask the supervisor how the pride of the school was progressing. The supervisor had in the meantime perused the finished pages of the candidate's script as they lay alongside him. The awe-spoken answer on one occasion during the arithmetic examination was somewhat as follows: " He is doing fine but he has made a bloomer in question X ". This was duly passed on in the hamlet and gloom ensued. However, Ernest was a fairly quick worker and he had time at the end to look over his answers to the early questions and duly discovered and rectified the error, with the result that he won the scholarship with record marks—Geography 125/130; History 125/130; English 130/140; Arithmetic 200/200; Total 580/600. Although Rutherford was about half a year older than the average of the other candidates, this success was remarkable when considered against the background of the one-teacher schools which he attended and in which he was unavoidably left much to his own devices. A survey carried out in 1924 by the author indicates that on the average the pupils of such schools at an age of 14 lag about one year in educational attainment behind those educated at larger town schools.

Thus Rutherford entered Nelson College in February 1887, being accompanied over the rough mountain forest track from Havelock to Nelson by his father, both on horseback. Because of his record and good grounding he was placed in the fifth form. In later years his then aged mother wrote to me an interesting story of the first interview she had with the Principal of the College, Ford, of which the following is an extract:—" After a short talk concerning Ernest and his abilities he said to us both: ' I have a personal favour to ask of you and will tell you the reason why. Bishop X's son is learning Greek as well as Latin, but where there is no competition there is no emulation, so I should esteem it a favour if you would allow your son to learn Greek as he has shown his knowledge of Latin in his exam. papers.' After thinking it over we consented and at the end of the year he won the Stafford and Senior Classical scholarships, so all was well."

The total number of boys at the College when Rutherford first entered was only 80, and it is little wonder that the relations between the masters and pupils were such that together they could go for ' hikes ' in the surrounding country and hills—hence the story of Littlejohn (the mathematics and science master) and Rutherford engaged in a geometrical discussion illustrated by rough markings with a stick on the soil beneath them.

Rutherford's period at Nelson College must have created deep, lasting and pleasant memories. Not only did he incorporate the name Nelson in his title, but within an hour of his death he said to his wife " I want to leave a hundred pounds to Nelson College. You can see to it", and then loudly "Remember, a hundred to Nelson College." He hardly spoke after that.

He was by no means a bookworm or what is colloquially termed a 'swot'. He entered into the full life and games of the school in every way. In the holidays he took his full share of household and garden chores and helped his father and eldest brother George in the sawmilling and flaxmilling operations. He fished in the Sounds, where in those days fish were even more plentiful than at present. He assisted the local road engineer with calculations of spoil from cuttings etc. He spent periods of his holiday with relatives earning pin money by hop-picking. It was on such a visit that he carved the potato-masher for his grandmother which is now held by the Royal Society. He was ever carefree and happy in his almost wholly outdoor occupations of work or play. His faculty of repartee, so evident in later life, was sharpened by rich healthy interplay with his equally lively and quick-witted brothers and sisters. He learned to cajole, manage and use them to help in his various jobs of work, but he also learned from his father the virtues of quiet purposeful solid accomplishment. Thus he had all the advantages of a country upbringing without the disadvantages of lack of mental stimulus, and this country environment was so much more primitively natural than that of the more densely populated England—there was so much majestic grandeur in the scenery. The maritime equability of temperature, the clear, clean atmosphere and strong sun, the forest surroundings, were all conducive to energetic action and joy of free adventure.

In touring the district with him in 1925 I remember well his sighs of disappointment that the scenery had been spoiled to little purpose by ruthless firing and destruction of so much of the bush. In his youth, unconsciously there had been stamped on his mind a lasting impression of something greater than that of man's creations, and he was longing to catch again some of its spirit, some of that peace and calmness of mind which comes from such contacts.

To return to his schooling, it was liberal, broad and not unduly specialized (as laid down in the Deed of Establishment of the College), although mathematics was probably his strong subject. Such was the distribution of marks in the University Entrance Scholarship Examination that only such a broad education would have enabled him to be successful as indeed he was in 1889, and to proceed to Canterbury College early in 1890.

In the meantime, the family had suffered two serious hardships. In the first place, his two younger brothers were drowned in a boat accident in the Sound. It was Ernest who, awed and shocked, ran home and blurted the news bluntly to the mother. The father and brothers scoured the shores for three months in a vain effort to find the bodies. This accident so seriously affected his mother that she for a long time lost her sunny, cheery nature, and never again did she turn to her favourite music or play again her cherished Broadwood piano. A little later, James Rutherford had a serious accident, fracturing five of his ribs. Shortly after his recovery, the Atkinson Government cancelled orders for railway sleepers, and the family perforce looked round for fresh avenues of occupation. His father crossed to the North Island and went North from Wellington on horseback, looking for suitable areas for flax. Eventually he arrived at Pungarehu, Taranaki, near the coast, 30 miles south of New Plymouth. Flaxmilling had not started in this area, so he was able to obtain suitable land at £3 per acre, near other flax swamps which he was afterwards able to cut under 'royalty'. Returning to Havelock he chartered the *Murray*, under Captain Vickerman, and loaded it with his whole family, three extra operatives, and all his household furniture, his horses, his flaxmilling machinery and a quantity of timber. The charter cost him £100. It took three days to get to New Plymouth, where the whole outfit was unloaded and the effects transported along the rough track to Pungarehu. Leaving his wife and younger children at New Plymouth, he began to carve out a home and establish his mill. Soon he was relatively prosperous, though his flax was sold in Melbourne at only £13 per ton and had to be transported to New Plymouth for shipment over a road which took nearly two days to travel, by a five-horse team drawing only three tons.

It is interesting to realize the energy and ability and powers of observation which James Rutherford put into his flaxmilling operations. He harnessed water-power to drive his mill. He experimented and developed a method of soaking the fibre after stripping and subsequently developed a special scraper to remove the vegetable matter so as to minimize the labour and time of paddocking. Much of his procedure was an adaptation of the cultural and hand methods used by the Maoris, which we seem to have now lost before extracting the scientific principles involved. He looked ahead and planted specially selected native varieties. Nevertheless, he relied mostly on ready grown swamp-flax, and such was the success of his operations that the flax he produced was reckoned amongst the best in the Dominion.

The move took place at about the same time as Ernest went to Canterbury University College and thus his University vacations were spent at Pungarehu. On one occasion he arrived at New Plymouth by boat from Christchurch, via Wellington, on a day when the coach south was not running, and without thought or hesitation tramped the 31 miles of rough road to the family home, which incidentally he took on the job of painting on one of his early vacations.

The early establishment and endowment of Canterbury College was another example of the importance attached to education by the early New Zealand Colonists in their determination to give to their children educational advantages comparable to those in the Old Country and available to a larger proportion of their children. Apart from a grounding in arts subjects and a smattering of biology and botany, Rutherford came under the influence of an able and conscientious Professor of Mathematics (C. H. H. Cook), and a stimulating but somewhat erratic Professor of Physics and Chemistry, A. W. Bickerton of the Royal School of Mines. Each did his teaching work conscientiously and well; Cook published mathematical papers relative to bridge structures in New Zealand and Bickerton speculated on astronomical subjects, his pet hobby, on which he wrote at length and often, being the study of stellar collisions and novae. In fact, one is tempted to wonder how far Rutherford was unconsciously influenced by these speculations in his later work on collisions of α particles with atomic nuclei.

Professor Eve and Dr. Feather have dealt ably with this period of Rutherford's training and there is little I would add, except to emphasize Bickerton's evidence that Rutherford was ever ready to assist his fellow students, and how in turn he was assisted by them. Rutherford became secretary of the College Science Society and a perusal of the minutes shows the extraordinary breadth of subjects of discussion, which appears remarkable from the viewpoint of these days of specialization. Rutherford's unconscious directness and humour crop up in the minutes from time to time. For instance, those of one of the meetings commenced, "It being cold in the normal lecture room, the Society adjourned to Mr. Page's room".

It was not until his fifth year, after graduating M.A., with a double first in Mathematics and Physics, and B.Sc. including Botany, that Rutherford commenced an original piece of research, and in this he could have had little assistance, other than stimulating enthusiasm, from his Professor. His subject of research related to the way in which a rapidly changing electric current is distributed in passing through a conductor, particularly a magnetic material such as iron. The preliminary experiments, as so often happens, clarified his ideas on the subject and led him to realize, as unknown to him Henry in America had realized, that a magnetized thin steel wire may be used as a detector of an oscillatory discharge, and the possibility of detecting Hertzian waves presented itself and he found himself able to send and receive signals over the length of the laboratory in 1894. Such an achievement was most creditable and the measurement of short periods of time and other experimental devices he evolved for his researches were most ingenious yet simple when considered against the background of the apparatus available and the isolation of having no peer with whom to discuss his work.

In passing, and to emphasize the nature of experimental work in those days, I may mention that he used as a source of electricity a battery of Grove cells whose zinc electrodes needed to be freshly cleaned and amalgamated each morning and the acid renewed; these were housed in a wooden box constructed at his Pungarehu home in the holidays.

It might be appropriate at this stage to mention that when the patents arising from the magnetic detector were under legal discussion at a later date the representative of a prominent firm of Patent Attorneys waited on Rutherford with a view to seeking his assistance as expert witness. In a letter to his principals on 5th November 1910, the representative wrote as follows:

"I beg to report herewith the result of my interview with Professor Rutherford on Tuesday, November 1st. Prof. Rutherford is most emphatic in his desire not to be associated with any side. He says that if he is retained or subpoenaed by either plaintiffs or defendants he would then be appearing in the interest of one side or the other. His position is one of complete detachment and disinterestedness and he feels that his independence would be surrendered if he assumes the position of expert witness, a rôle to which he is averse.

"Professor Rutherford tells me that he has never exploited his scientific work for commercial purposes, or with a view to patenting, and thereby securing for himself alone the benefit of such research. He fully and freely publishes the result of all his scientific work and therefore his research possesses the greater value in as much as it is not done behind closed doors, but it is done with a view to adding to the world's knowledge."

Rutherford used the account of his magnetic work in his application for the 1851 Exhibition Scholarship, and also later as a basis for further research at Cambridge in 1895. At first the New Zealand University Authorities recommended that the scholarship be given to a chemist, J. C. Maclaurin. It is always difficult to assess the relative merits of candidates for this scholarship who present work from the various Colleges and in different sciences. Fortuitous factors arise which determine the choice of candidates. Maclaurin was certainly a fine man and a most able chemist, who afterwards filled with distinction the post of Chief Government Chemist of New Zealand. He was the brother of the R. C. Maclaurin who later was one of Rutherford's companions at Cambridge and in due course President of M.I.T. at Boston where he laid the foundations for its present eminence. Rutherford, thinking he had missed the award, took a temporary teaching post at a secondary school. He had also previously entered into correspondence with a college friend, Stevenson, who had gone to Edinburgh, with a view to entering on a medical course there as one way of obtaining overseas training and experience, which is so eagerly longed for by those in relatively isolated New Zealand, and which indeed is even now robbing her of her brightest potential leaders. However, in the meantime Maclaurin had married and accepted a position as Government Analyst and thus was unable to take up the scholarship, so that it was offered to Rutherford. The scholarship was of a value of £150 per annum for two or three years and there was no allowance for the expenses of passage to the U.K., so Rutherford perforce had to borrow, and this necessitated cheap and careful living for the first few years of his career in England.

I will now pass quickly over his subsequent experiences at Cambridge with Sir J. J. Thomson, and at Montreal. At Cambridge he at first pushed forward his work on the reception of Hertzian waves until it was reasonably rounded off with work which provided material for a lecture at the British Association, and a paper in the *Philosophical Transactions of the Royal Society* which gave him some prestige and standing. He then wisely threw in his lot with J. J. Thomson and the main work of the Cavendish Laboratory, particularly under the stimulus of the sensational discovery of x-rays and of their remarkable properties. 'J. J.' had discovered that x-rays enabled electricity to pass through a gas by a process resembling that of the passage of current through an electrolyte. Rutherford's first note on the subject seized on the important point that the current between two plates separated by gas exposed to x-rays increased with the distance between the plates even though the potential gradient was less.

Rutherford assisted Thomson, and himself carried out a series of ingenious experiments which established the main principles of the ionization theory of conduction of electricity through gases as we now know it. From this he was led to examine the ionizing effect of the radiations from uranium, and his main life's work on radiaoctivity then started. He had only proceeded as far as showing that the ionization produced in gases by uranium rays was of the same nature as that produced by x-rays when he transferred to McGill University, Montreal, as research professor at £500 per annum, and thenceforward he plunged immediately into a brilliant series of experiments in which he was himself the leader and inspirer of a group of workers who gradually gathered round him—and whose previous vocations had been as electrical engineers, chemists or physicists. Thenceforward he was the pioneer, planning each advance, consolidating knowledge won, integrating the efforts of his colleagues and developing bold theories to guide subsequent work. In succession he discovered that the uranium radiation was complex—he named the two major ionizing components alpha and beta. He proved that the alpha radiation was particulate and positively charged, and deduced that it consisted of charged helium atoms. He discovered thorium emanation in a series of experiments described by Sir Henry Tizard as the finest of his career, and unravelled the nature of series of radioactive changes with subsequent products giving particular kinds of radiations, and associated these definitely with the energy emitted, which he measured in the form of heat. All this work was consolidated in his Bakerian Lecture to the Royal Society in 1904, which finally placed the facts and theory of radioactive transformation on a universally accepted basis.

We now come to the Manchester period of Rutherford's work. Schuster had written offering to retire from the Professorship at Manchester if Rutherford would take his place. The conditions were such that Rutherford would have a minimum of lecturing duties. Schuster also provided the finance for a mathematical readership which was held in turn by Batemen, Bohr and Darwin, all of whom were later to contribute to Rutherford's field of work. Rutherford arrived at Manchester about three months before the session, which started at the end of September 1907, and I well remember a sort of inaugural discussion and meeting at the Physical Colloquium with A. Schuster, J. J. Thomson and Rutherford all present, three giants all beaming and smiling in happy reunion.

I was then a callow youth in the second year of my honours course. The arrival of Rutherford decided my fate—and what a spur to energy he was! I quickly ran through all the practical exercises in electricity and magnetism and optics etc., and then proceeded as the first of the guinea pigs in those beautiful elementary experiments in radioactivity drawn up by Rutherford and Geiger, and afterwards published by Makower and Geiger. We constructed our own electroscopes and obtained first-hand acquaintance with the absorption and other properties of the alpha, beta and gamma rays, and with the radioactive transformations of radium emanation and its products—and similarly with thorium and actinium.

Soon I approached the frontier of new knowledge, it did not take so long in those days, and I was allowed to help Rutherford in experiments on phosphorescence produced by α and β rays and later to act as Geiger's assistant in experiments on the scattering of α particles, compound scattering as we later termed it, and on the counting of α particles using the method, then under development, of magnification of the effect through ionization by collision and using a Dolezaleck electrometer. Geiger was a master of this instrument, which needed much skill if accurate results were to be obtained. I was a member of the O.T.C. and one Saturday morning when working in uniform preparatory to an afternoon's military exercise, I happened in sheer admiration to say, "Dr. Geiger you *do* know your electrometer." His reply was, "Ah, but I know my gun better than I know my electrometer". Although he was a Frank and not a Prussian, such was the effect of conscription. Alas, Geiger was to fight against us as an artillery officer in the war which followed 5 years later.

Geiger was a delightful man, energetic, orderly, kindly and methodical, and he was most loyal to the Professor. We tried to teach each other our respective languages. He read Eddington to me and I read Planck's *Thermodynamics* to him and we tried to correct each other. I hope I did not give him too much of a Lancashire accent.

Rutherford was also engaged with Royds in that beautiful experiment on the spectrum of the radium emanation and production of helium from α particles fired through a thin-walled container of radium emanation, which showed definitely that the α particles were helium. I remember the details so well because the spectroscope was set up in the same room as that in which I worked on phosphorescence. Photographs of the spectrum of radium emanation and helium were taken at intervals as the emanation decayed and the helium spectrum appeared. One day someone had been too inquisitive in looking at the spectroscope. Rutherford came into the room and, noticing that the prism had been displaced, flew into a towering rage. He came over to the optical bench at which I was working and placed his hand round the back of my neck, only moderately gently, and said, "Did you move that prism?" I knew and trusted him too well to have the slightest fear of him and answered "No". I was sufficient of a school boy to enjoy the 'boss' in a tantrum. Half an hour afterwards he came back to the room and sat deliberately on a stool alongside me and quietly expressed his apology for getting his 'dander' up and accusing me of the misdemeanour. He must have found out the culprit in the meantime. Then we went on to one of those helpful and, on his part, non-condescending discussions of the progress of my work, which all who had a similar privilege treasure as their happiest recollections. I have never known him to bear malice. You will appreciate his action when I add that I was only 19 years of age at the time—in fact I realize how extraordinarily fortunate I was to work under his aegis and to make two contributions to the Royal Society and be allowed to read one of them myself, before I had passed the age of 20.

One day when I was exercising my privilege of working with Geiger, Rutherford came in and a discussion ensued between them as to the nature of the huge electric or magnetic forces which could turn aside or scatter a beam of α particles when passing through such a thin film of gold (some 10^9 gauss). Rutherford turned to me and said, "What about trying whether you can get α particles reflected from a solid metal surface". I don't think he expected any such result, but it was one of those 'hunches' that perhaps some effect might be observed, and that in any case that neighbouring territory might be explored by reconnaissance. Rutherford was ever ready to meet the unexpected and exploit it, where favourable, but he also knew when to stop on such excursions. Naturally, I knew enough to appreciate that even though a negative effect might be expected yet if I missed any possible result it would be an unforgivable sin. Accordingly I made quite sure of preparing as strong a source of radium emanation as I possibly could in a conical tube closed by a mica window—a technique which had been developed by Geiger. To my surprise I was able to observe the effect looked for and I collected reflectors of metals from aluminium to platinum and made comparative measurements. I remember well reporting the result to Rutherford a week after, when I met him on the steps leading to his private room, and the joy with which I told him that the effect seemed to vary approximately as the 3/2 power of the atomic weight and not as the square root which was the result obtained by Geiger in the work on compound scattering. Unfortunately I had to report an exception—silver. I had borrowed a coin of allegedly pure silver from a Russian scientist (Antonoff) working on electrochemical effects, and at the time did not realize that he had contaminated the surface with polonium! A few weeks later Rutherford instructed that I should round off the experiment with Geiger in a form suitable for publication.

Rutherford thought over these remarkable results for many weeks. Such ponderings and related speculations and calculations were generally done in the quiet of his study at home, but out of it emerged his inspiration of the nuclear or planetary atom with its features of resemblance to a solar system in miniature, a brave conception in view of its reversal of the ideas current at the time, based on those of J. J. Thomson (1910) and Kelvin (1902) and in which the positive electricity of atoms was considered as dispersed in a relatively large sphere instead of being concentrated in a relatively small nucleus. The solution was a direct answer to the problem he set himself to explain, i.e. the origin of the huge forces necesssary to deflect the fast-moving α particles.

The dawn of the solution has been recorded by Sir Charles Darwin in a letter as follows:—

> "I count it one of the great occurrences of my life that I was actually present half an hour after the nucleus was born. It was a Sunday supper at his Manchester house and I remember him saying to us that he had been looking into the big scattering of α particles and that there must be enormous forces in the atom to do it. Then he said it was a charge of something like a 100 electrons to do it in gold.
>
> "The other thing I remember is that he was at once deeply interested in the distance of closest approach, thus showing that he wanted to get on to the next problem but one even before he had proved the next result."

Rutherford made all the mathematical calculations himself, though he later asked Darwin to verify them, and also to check by working out what would happen in a possible magnetic analogy of a large force approximating with distance to the inverse cube rather than the inverse square. Rutherford deduced the relation of the extent or probability of 'single' scattering through different angles from atoms of different nuclear charge, and also the relation between the velocity of the α particles and the probability of their being scattered. In addition he deduced from our results that the positive charge on the nucleus had a numerical value approximating to half the atomic weight.

Preliminary checks of the former were made by Geiger over a limited range of angles, and at that stage Rutherford was sufficiently confident to publish the whole story, and this announcement was received with the same bewildered astonishment as so many of his previous major pronouncements, and again the outstanding feature was the simplicity of the solution. To check the theory completely was a large task. In the meantime I had accepted an appointment at Queen Mary College with Professor Lees and been fortunate enough with the assistance of Barrett to work out the probability law of distribution of time intervals betweeen successive disintegrations, with particular reference to uranium transformations, and also to discover the branch disintegration of thorium C. Rutherford kindly invited and made it possible for me to return to Manchester to undertake experiments with Geiger on the more detailed tests and consequences of the theory.

The complete check was a laborious but exciting task. I remember Geiger making a calculation that in the process of the work we counted over a million individual α particles. When I look back and consider the apparatus used and the nearness of our heads and bodies to the large sources of radium emanation used and the time of exposure to the radiation, I marvel that it did so little physical harm to us. On modern safety standards and using the same method of counting scintillations the apparatus would need to have been almost unworkably complicated for the purpose.

Our work did, I think, sufficiently establish the theory on a firm basis. We had checked the effect with angle over a range of 1 : 250,000, with velocity 1 : 10 and with atoms from carbon to platinum, all the results being in accordance with theoretical expectation. Fortunately, within 2 years, Moseley's experiments gave a more precise significance, in terms of the order of elements in the periodic table, to the effective number of positive charges in the nucleus of successive atoms, while Niels Bohr, using considerations of quanta, provided an answer to the difficulty of explaining the production of spectra from the atom model. It may be mentioned in passing that Rutherford's determination of the value of **e** two years earlier as approximating closely to that deduced by Planck from a general optical theory of natural temperature radiation, had in the meantime also directed increased attention to the significance of Planck's quantum theory as applied to electromagnetic radiation in general.

The nuclear atom was thus an outstanding example of Rutherford's boldness of conception and simple directness of attack on the problem of explaining a definite experimental result. It is true that Nagaoka had speculated on some such form of atom in 1904, a Saturnian atom with rings of rotating electrons, but this fact was only brought to Rutherford's notice by the elder Bragg in subsequent correspondence. I think that throughout it was Rutherford's powers of intuition and directness which enabled him to visualize the picture of his α particles in their approach to atoms of the scattering material unhampered by undue abstruse mathematical conceptions; it was akin to Bickerton's more speculative methods on less evidence in considering stellar impacts in the old days when Rutherford was a student at Christchurch. The difference was that Rutherford conceived the deflections of α particles as due to repulsive forces instead of those of attraction.

While the theoretical consequences of the nuclear atom were being explored and its application extended, I had amused and interested myself in general work on branch disintegration and passage of α particles through matter etc.

I would like to interpose at this stage a few words on the spirit of the team of workers under Rutherford at Manchester at that time. The outstanding feature was the spirit of harmony—each knew what the others were working on, each gave ideas freely to the other, no one was secretive. It seemed almost immaterial who did the work and published the results. There were good pickings for everyone and no one jostled for a particularly spicy titbit or line of work. So often an idea which arose spontaneously at the afternoon tea discussion would be taken up by one or generally a pair of workers who happened to have opportunity at hand. There was of course the overriding, shrewd, guiding but tolerant hand of Rutherford, who watched the general front of attack. It would be invidious perhaps to give examples but these can be found in the free acknowledgments given at the end of each of the large output of papers published. It is noteworthy how many of the papers are of joint authorship. It seemed as though the natural unit for experimentation was two. There was ever present that warmth, glow, cheerfulness and courage, originating from Rutherford himself but with all those working in the laboratory acting as secondary radiators, which can so greatly strengthen and fortify cold effort and efficiency. Leadership was not only or so much impressed from above as integrated from all levels, and thereby we were all trained to think constructively. Rutherford was of course a 'tiger for work' and he expected the same from all, but as he himself wrote to Geiger in later years, "They were happy days in Manchester and we wrought better than we knew".

To return to the question of encounters of α particles with the nuclei of atoms, consideration has so far been limited to heavier atoms from carbon to platinum, i.e. to cases where the recoil of the nucleus did not affect significantly the experimental results. Moreover, in the case of gold, for example, it would be calculated that the nearest approach of α particles and nucleus was of the approximate order 3×10^{-12} cm., which was of the order of 10 times the accepted value of the diameter of the electron. It was generally accepted that the whole of the mass of the electron was probably electrical in origin but at the time it was generally thought that positive electricity somehow was incorporated with gravitational mass. However, Rutherford and Darwin calculated that in the collision of an α particle with a hydrogen nucleus of unit positive charge the nearest approach could be about $1{\cdot}7 \times 10^{-13}$, assuming both acted as the equivalent of point charges and that the inverse square law of force still held. As this distance is less than the diameter of an electron it raised the question as to whether, if the hydrogen nucleus had a radius of some 1/1,830 of the electron, its mass could be explained as electrical. Thus it became of interest to investigate the collisions of α particles with 'H' nuclei in as direct a manner as possible.

Elementary consideration of a head-on collision of an α particle of mass 4 with a hydrogen nucleus of mass 1 showed that the latter should take on a velocity of 1·6 times that of the incident α particle, and presuming that Bragg's Law of slowing down or absorption could be applied and allowing that it could be presumed to be singly charged, it should have a range of approximately four times that of the incident α particles. Darwin worked out the general case of oblique incidence.

In 1914, therefore, I decided, naturally with encouragement from Rutherford, to try the experiment, at first in a simple way, and in general the results proved to be largely as anticipated, i.e. the 'H' particles resulting from collision had a range of approximately 100 cm. in hydrogen, and the absorption in metal foils from aluminium to platinum proved to be approximately as Bragg's Law, i.e. according to the square root of the atomic weight of the absorbing metal. The velocity of 'H' particles arising from oblique incidence was found to be smaller than those arising from direct impact. At this stage an attempt was made to test Darwin's calculation that the numbers of 'H' particles projected at an angle of θ to the direction of the incident α particles should vary as $\sec^3\theta$ because of the importance of such measurements to the questions of size and shape of the colliding nuclei. An intense parallel beam of α particles was fired into a film of wax of approximate composition $C_{27}H_{56}$ and observations made at various angles. A serious anomaly showed itself. Far too many scintillation H particles were observed and some of these were traced to the source of radium emanation itself. I spent much time and trouble in efforts to exclude any possibility of hydrogen contamination of the source, but in all cases, even with a quartz x-ray tube or RaC deposited on nickel, one could in air observe H particles at well over 10 cm. from the source.

At this stage I was appointed to a position in New Zealand and also war broke out and the experiments were hurriedly written up in a second paper on the subject, which ended "There seems a strong suspicion that H particles are emitted from the radioactive atoms themselves, though not with uniform velocity". It was impossible for me to continue the experiments in New Zealand, and shortly afterwards I returned to active service in France to join the rest of my ex-colleagues from the laboratory.

Rutherford was extremely interested in the results, however, both for the reasons already stated and also because the emission of no light element other than helium had been previously observed in radioactive transformations. Characteristic of his selflessness he wrote asking if I *minded* if he took over the work, and during the next four years, in such intervals as he could spare from his war activities, mainly in connection with problems of submarine detection, he repeated and extended the experiments. Although he agreed with the possibility that a few H particles did arise from the radioactive source, he found that the majority of the particles observed in some of my experiments arose in a very different and surprising manner, i.e. from actual disintegration of nitrogen in the air following the effects of intimate collisions of the alpha particles with nitrogen nuclei.

In considering the work of Rutherford in this period, assisted only by the laboratory steward, Mr. Key, the difficulties of making accurate observations of such feeble scintillations, using the microscopes then available, and the tremendous complexity of the radiations which he found to be involved, one cannot but be filled with admiration of his pertinacity, his logical sequence of experiments, his marvellous intuition in making deductions from the results of each experiment, and his boldness and directness of attack on each clue presented. He found that nuclei of atoms such as oxygen and nitrogen could be set in sufficiently rapid motion by α particles, so that they themselves could produce scintillations on a zinc sulphide screen. He established by magnetic and electrical deflection that the particles observed in hydrogen were actual hydrogen nuclei. Of more importance, however, were his deductions regarding the rapid changes of the nature of the forces between an α particle and nucleus of a light atom, when the apsidal distance was reduced to about $3{\cdot}5 \times 10^{-13}$ cm., indicative of asymmetrical shape. The interforces at this distance are of the order of 5 kg. weight, and not only was there evidence of the asymmetrical shape of the α particle but there was possible evidence of deformation of the particle under these circumstances, but not sufficient to break up the helium nucleus into constituent parts.

The crowning achievement, however, of a logical series of experiments, was the observation that in the case of collisions of an α particle with nitrogen there was a definite anomaly as compared with similar collisions with carbon or oxygen, and that "we must conclude that the nitrogen atom is disintegrated under the intense forces developed in a close collision and that the hydrogen atom which is liberated formed a constituent part of the nitrogen nucleus".

Rutherford enjoyed these experiments; as he himself said later, "I know of no more enthralling adventure than this voyage of discovery into the almost unexplained world of the atomic nucleus". It is remarkable that Rutherford made so few errors in his speculations on the results of these various experiments, in fact to mention one or two examples only serves to emphasize the overall boldness of his deductions. He had been very concerned because apparently there was an undue proportion of 'double scintillations' as evidence of possible emission of two H particles at once from some of the atomic collisions. I was privileged to spend a month with him at the conclusion of the war while awaiting transportation to New Zealand, to make check on some of these questions before publication. With such faint scintillations as were involved, and particularly as the field of view extended over several degrees, it is difficult to estimate small time intervals between the appearance of scintillations at various parts of the field, and it was found that scintillations appearing at 1/10 second intervals were apt to be recorded as simultaneous doubles. It is of interest to note that in the original observations of the counting of individual α particles by Rutherford and Geiger, a similar misinterpretation was made, but due in this case to the long period of the needle of the Dolezaleck electrometer and the consequent loss of time resolution. Thus they recorded, in the paper referred to, a curve showing the distribution of time intervals between successive α particles, which approximated nearer to a Gaussian distribution than to the experimental curve established later.

Similarly, Rutherford deduced the probability of formation of an atom of atomic weight 3, which on further investigation proved to be unlikely. Nevertheless, his remarkable experimental evidence and deduction of artificial disintegration of nitrogen was soundly based and was verified a few years later by Blackett in cloud trail photographs, which also indicated that the α particle was actually first absorbed into the nitrogen nucleus, producing an unstable atom which immediately exploded with emission of the H particle observed. I may add in passing that at a later date Rutherford suggested that the H nucleus and H particle be termed the Proton because it sounded to him somewhat akin to Prouton.

Thus these experiments, made by Rutherford under such trying circumstances and with simple, cheap apparatus, were the pioneer observations leading to the accomplishment by Chadwick, Cockcroft and others, of the disintegration of a whole series of atoms and ultimately to the utilization of the energy liberated. Indeed the last sentence of the original paper in 1919 on the production of hydrogen from nitrogen is prophetic, and reads *: "The results as a whole suggest that, if α particles—or similar particles—of still greater energy were available for experiments, we might expect to break down the structure of many of the lighter atoms".

In the foregoing I have tried, I hope objectively, to indicate that for much of Rutherford's methods in his life work we are indebted to the influence of his parents and of the home life and environment in which he was raised, i.e. his mental equipment, his sterling character, his abounding energy and enthusiasm, his direct approach to the problems confronting him, and above all his human characteristics which enabled him to attract and retain a happy and willing band of co-workers. I feel that we need to take note of these things in their relation to desirable features in the development of our educational and social systems since the greatest need of our age is the production of inspiring leaders, of which he was the outstanding example of our generation.

As further evidence I can but conclude with the words of the noble tribute of Sir James Jeans, given at the time of his reading of Rutherford's Presidential address to the Indian Science Conference in January 1938, which had been prepared by Rutherford about two months before his untimely death:

> "Those of us who were honoured by his friendship know that his greatness as a scientist was matched by his greatness as a man. We remember, and always shall remember, with affection his big, energetic, exuberant personality, the simplicity, sincerity, and transparent honesty of his character, and, perhaps most of all, his genius for friendship and good comradeship.
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> In his flair for the right line of approach to a problem, as well as in the simple directness of his methods of attack, he often reminds us of Faraday, but he had two great advantages which Faraday did not possess—first, exuberant bodily health and energy, and second, the opportunity and capacity to direct a band of enthusiastic co-workers. Great though Faraday's output of work was, it seems to me that to match Rutherford's work in quantity, as well as in quality, we must go back to Newton. In some respects he was more fortunate than Newton. Rutherford was ever the happy warrior—happy in his work, happy in its outcome, and happy in its human contact."

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* *Phil. Mag.*, 1919, **37**, 587.

A New Theory of Recrystallization Nuclei

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MS. received 16*th June* 1949 ; *read* 3*rd February* 1950

ABSTRACT. After a review of the numerous experimental observations in this field which need to be explained, a new theory is presented of nucleus formation when a cold-worked metal is annealed. The nuclei are held to be formed in the most distorted parts of the lattice, in the 'local curvatures' postulated by Burgers. At these places the lattice locally regains its perfection by the recently discovered process of diffusion of dislocations, or 'polygonization', and the perfect nucleus so created is then able to consume the less perfect crystal in its vicinity. Experimental evidence for this picture is presented. Each potential nucleus is held to have a definite incubation period (as distinct from the fixed probability of becoming active which was the basis of previous theories). By means of certain assumptions as to the distribution of curvatures and the dependence of incubation time on curvature, a quantitative theory is developed, which is found to be capable of accounting for all the experimental data on nucleation rates. The paper concludes by comparing critically the new theory with older ones.

§ 1. INTRODUCTION

In the last twenty-five years there has appeared a wealth of experimental results bearing on the recrystallization of deformed crystalline solids. It was early recognized that the process took place by the *growth* of new crystals from *nuclei* originating in the deformed matrix; and more recently it has become clear that for a better understanding it is necessary to study the kinetics of the process, in particular the two basic rates, the rate of nucleation N and that of growth of the nuclei once they are formed, G. Important articles on this subject are those by Kornfeld *et al.* (1934, 1935, 1937), Johnson and Mehl (1939), Stanley and Mehl (1939), and Anderson and Mehl (1945). The last of these studies is by far the most thorough. Polycrystalline high purity aluminium was used, and the effects of time, temperature, deformation and grain-size on both N and G were investigated. Here N was the number of nuclei formed per second per cm^3 of *un-recrystallized* material, and G the rate of increase of diameter of the approximately circular grains which were formed in the thin sheet specimens used. G was calculated from direct measurements of numerous individual grains, and N was computed as a function of time by extrapolating back for each grain to the instant when its diameter was zero. The present paper is an attempt to explain some of the results obtained by Anderson and Mehl in terms of a quantitative model of the nucleation process. No attempt will be made to deal theoretically with G, which appears to be physically quite distinct from N.

The chief results emerging from Anderson and Mehl's work which require explanation, are:

(i) The rate of nucleation in weakly deformed polycrystalline aluminium is not independent of time. It is zero at first (i.e. there is an induction period) and then increases. The shape of the plot of N against time is approximately exponential at first, but at a later stage a point of inflection is reached if the curve can be followed far enough. The end of the curve occurs when the whole of the deformed matrix has been consumed by the new grains.

(ii) In weakly deformed metal of coarse grain size there is a period during which N increases sharply, followed by a decrease in N sharp at first and then more gradual. The values of N are smaller and the point of inflection occurs after a considerably longer time than for fine-grained metal similarly deformed.

(iii) The same shape of curve is reported for heavily deformed fine-grained metal; sometimes N decreases from the first value plotted.

(iv) The whole curve described in (i) is shifted towards higher values of N and shorter times as the temperature or the deformation increases.

In addition, there are a number of observations due to other workers which any theory of nucleation should be capable of explaining. In the first place, the evidence is overwhelming that nuclei are formed in regions where the damaging effect of deformation is most marked. Nuclei appear preferentially in active glide-planes and near curved twin-bands, and the concentration of new grains at or near crystal boundaries is particularly clear. This last fact is significant since it has been proved beyond doubt that there is a sharp shear *gradient* in these regions, a state of affairs which must be associated with considerable inhomogeneity of the lattice. The point is raised again below.

Secondly, it has been found that there is a statistical connection between the orientation of the deformed crystals and those of the new grains. This is true both for single crystals and for polycrystalline aggregates (Burgers *et al.* 1928, 1931).

Finally, it seems that a 'recovery anneal' at a temperature too low to cause recrystallization leads to an increase in N in the case of single crystals (Kornfeld and Shamarin 1937). This has recently been confirmed by Burgers (private communication). When polycrystalline specimens are used, both a retardation (Kornfeld and Pavlov 1934, Stanley and Mehl 1939) and no change (Anderson and Mehl 1945, Eastwood, James and Bell 1939) have been recorded.

§ 2. NATURE OF RECRYSTALLIZATION NUCLEI

As has been mentioned in the first section, recrystallization nuclei form in certain distinctive parts of the structure of a cold-worked metal, and therefore we must begin by examining the cold-worked state. Microscopic and x-ray evidence combine to show that the lattice of a crystal is considerably less perfect after cold work than it was before. X-ray patterns show that the lattice becomes curved, which is a sign of the presence of internal strains. These strains would seem to be predominantly on a comparatively coarse scale; this follows both from x-ray work and from the microscopic observation, by many authors, of *deformation bands* in cold-worked grains. The lattice in these bands has been shown conclusively to be rotated relative to the rest of the grains, often in a complicated way. Experiments by the present author (in course of publication) suggest that these bands are associated with uneven yielding of different parts of a grain, and are the remains of flexural glide at the limits of those parts of the grain which have yielded first.

There is, however, another type of internal strain, which is much more highly localized than the deformation bands. This type has been called 'local curvature' by Burgers (1935); it occurs in the vicinity of the glide zones and is the necessary consequence of the interaction of dislocation of opposite signs which become immobilized in large numbers on these zones and lead to work-hardening (Taylor 1934). Each glide zone consists of a large number of separate active glide-planes, as has recently been shown by Heidenreich and Shockley (1948). Many units of slip take place on each of these active planes, but eventually they 'seize up' and slip spreads to the neighbouring plane. According to the Taylor theory of work-hardening, such seizing up must be due to the interaction of dislocations of opposite signs. In some places this interaction seems to lead to a catastrophic piling up of dislocations, and the result is a local curvature, as sketched in the upper half of Figure 1. It is particularly likely to occur in places where there is a marked shear gradient, such as the boundary regions in polycrystals where many microscopically visible glide zones fade out and disappear. Another type of place where this seems to happen is in the deformation bands mentioned above. Here glide zones stop quite abruptly, and sometimes one finds individual glide zones which are bent much more sharply than their neighbours. Figure 2 (see Plate) is a photomicrograph showing such a place at high magnification; the resemblance to Figure 1 (*a*) is clear. There is some direct evidence for the existence of these localized strains, though in the writer's opinion the best evidence is indirect. They are a necessary consequence of the dislocation theory of cold work, and this theory in turn has received so much experimental verification that it is now widely accepted. The direct evidence is mostly microscopic. Thus, McLean (1947) and Burke and Barrett (1948) have both shown that when brass is heavily cold-worked, and the specimen is then sectioned, polished and etched, the glide zones show up quite clearly. This must mean that the lattice inside the zones had been disturbed to such a degree that it is preferentially etched. Again, the disturbed state of the glide zones was made clear by Berg (1934) and Barrett (1945) by the use of the technique of x-ray microdiffraction developed by them. The evidence given by x-ray Laue photographs is ambiguous, and has given rise to protracted arguments as to whether the internal strains revealed by asterism are due to large-scale strains (i.e. deformation bands) or to local curvatures, the implication being that one or the other must be the case, but not both. Without going into the controversy in detail, it may be said that it seems very probable now that Laue asterism is always the consequence of large-scale curvatures of the lattice; but that, wherever these occur, they are accompanied by local curvatures, which probably occupy too small a fraction of the lattice to affect the x-ray pattern. Even if they do affect it, this would be masked by the asterism due to the coarse curvatures.

Several years ago Burgers and Louwerse (1931) suggested that nuclei were formed in the local curvatures when a deformed crystal was heated. It was argued that the Laue asterisms were due to local curvatures. When a crystal was heavily compressed and then recrystallized, the new grains fell into groups of orientation rotated about the same axis relative to the mother crystal as the curvatures deduced from the asterism, though it was necessary to assume that the range of rotation actually present was greater than that calculated from the Laue photographs. It has now been suggested that the asterisms are due to coarse curvatures; but Burgers' argument remains valid because the axes of curvature of both the coarse and the local curvatures would be expected to be the same, according to the theory of dislocations. He had to postulate that the new grains kept the orientation of the lattice in the local curvatures, without being able to describe a mechanism by which they might do so, the difficulty being that the local curvatures would have to become strain-free before they could grow. Such a mechanism has recently been discovered by Cahn (1948, 1949) and by

Guinier and Tennevin (1948). It is known as *polygonization*, as it consists of the change of a continuously bent lattice into a number of polygon elements or lamellae, each keeping the orientation of that part of the bent lattice from which it has been formed, but free of elastic strain. Figure 3 (see Plate) is a photomicrograph of an aluminium single crystal which has been bent and then polygonized by annealing. The boundaries between the lamellae have been revealed by the special etchant developed by Lacombe and Beaujard (1947) and show up as rows of etch-pits. After etching the crystal was bent a little further to develop fresh slip-lines (which are faint because the section is almost parallel to the slip-direction); these serve to show that the boundaries are perpendicular to the slip-planes. Figure 4 shows schematically the state of the lattice of the crystal before and after annealing. In fact the angles between the

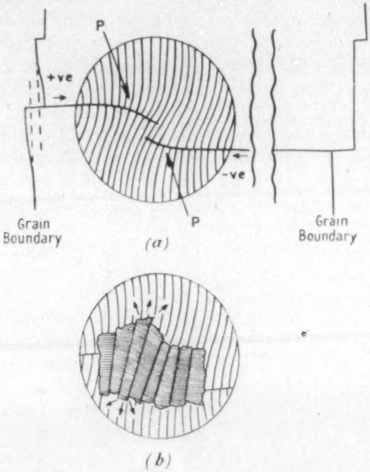

Figure 1. Local curvature in a deformed crystal: (*a*) as deformed; (*b*) after annealing.

lamellae are only a few minutes of arc and no discontinuity in the direction of the slip-lines is therefore visible in Figure 3. In the original papers it is argued from the fact that the boundaries are perpendicular to the glide-plane, and from other facts, that the process takes place by the diffusion of dislocations through the bent lattice. The lattice is thus relieved of elastic strain, while the dislocations collect along surfaces, separating lamellae inclined slightly to each other (see also the full discussion by Cottrell 1949). When the annealing is much prolonged, some of the parallel-sided blocks seen in Figure 3 grow at the expense of the others and become very large. It has also been found (Cahn, in course of publication) that polygonization can occur, not only in freely bent crystals, but also within deformation bands in extended crystals; the blocks are formed within the bands, and then some of them tend to swallow their neighbours and spread into the

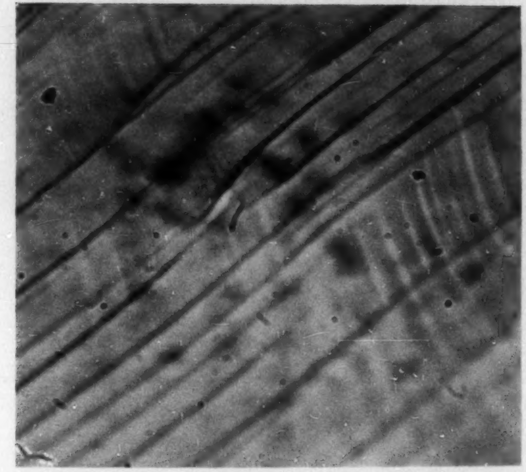

Figure 2. Local curvature in a deformation band. Aluminium crystal, extended 30%. (Magnification ×2,300.)

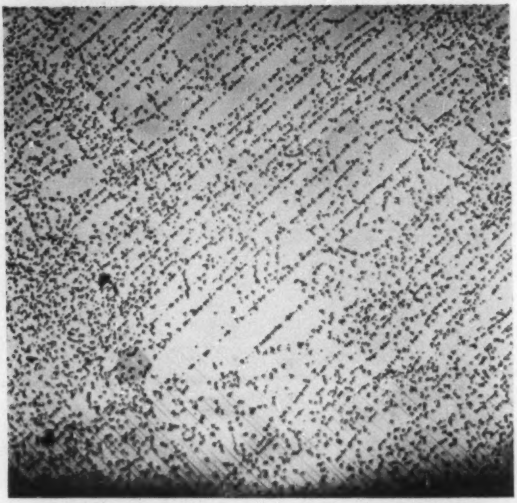

Figure 3. Polygonization boundaries in a bent and annealed aluminium crystal. (Section almost parallel to glide direction.) (Magnification ×90.)

adjacent lattice. It is now suggested that the same thing can happen on a still smaller scale, and that local curvatures of the kind discussed above can become polygonized; moreover, after a time one of the blocks grows at the expense of the others and then grows outwards and begins to consume the surrounding cold-worked metal. On this view, then, a recrystallization nucleus is a polygonized local curvature. The lower half of Figure 1 shows schematically what is meant. It will be seen how this model satisfies Burgers' postulate that the growing grain retains the orientation of the local curvature, and gives a physical basis to his explanation of the preferred orientations obtained by recrystallization.

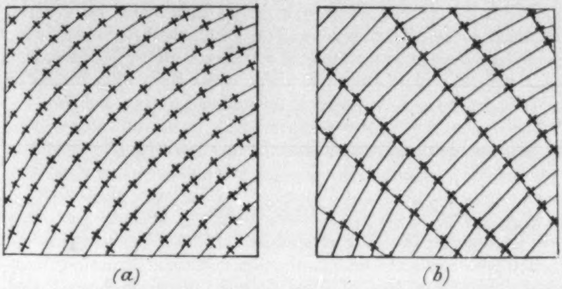

Figure 4. Nature of polygonization in a bent crystal lattice: (*a*) as bent; (*b*) annealed. The crosses represent *excess* positive dislocations remaining on the glide-planes after bending.

§ 3. KINETICS OF NUCLEATION

An attempt is made in this section to develop the model of nucleation just put forward in a more quantitative way. For this to be done, it is necessary to make certain assumptions. The main assumption is that the time necessary for a local curvature to become polygonized sufficiently at a given temperature for growth to start (i.e. for it to become an active as distinct from a potential nucleus) is the shorter the more sharply it is bent, i.e. the smaller the radius of curvature. Experiment indicates that the initial stages of polygonization do not take place more quickly in a sharply bent crystal than in a weakly bent one, but that the later stages, where a few blocks grow at the expense of the others, thus rendering them visible on an ordinary Laue photograph, come sooner in a sharply bent crystal.* It is believed that this stage must in fact be reached before the nucleus becomes active. This belief is founded on the principle, generally valid in processes of nucleation and growth, that large nuclei of a second phase can grow more readily than small ones. Moreover, in the present case the nucleus will not be able to start growing until a block has ruptured its continuity with the parent lattice (compare the somewhat analogous case in age-hardening), and this can only occur when a block has grown large enough for there to be a considerable stress where it meets the parent lattice.

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* Moreover, microscopic study has shown that this last stage is reached much more rapidly in the polygonization of deformation bands (where the radius of curvature is a small fraction of a millimetre) than with a microscopically bent crystal.

In detail the assumptions are as follows:

(i) The curvature of each potential nucleus can be expressed in terms of a single radius ρ. For the present purposes this may be taken as the smallest radius present (at P in Figure 1).

(ii) The curvatures $1/\rho$ of the potential nuclei are normally distributed about a mean $1/\rho_0=\alpha_0$, characteristic of the nature and degree of deformation and increasing with the latter. Different arbitrary values may be assigned to the mean deviation σ of the distribution and the results compared; it is found that they are not very sensitive to the values of σ chosen. Since deformation is always microscopically very inhomogeneous, there must be a large mean deviation. The theory is worked out for values of σ equal to $\frac{1}{3}\alpha_0$, $\frac{1}{2}\alpha_0$, and $\frac{3}{4}\alpha_0$.

(iii) The time t taken by a potential nucleus to become active is a function only of $1/\rho$. Two assumptions are made in turn (writing α for $1/\rho$):

$$\text{I.}\quad t=\tau\rho=\tau/\alpha\,,$$

$$\text{II.}\quad t=\tau\rho^2=\tau/\alpha^2,$$

where τ is constant for any one temperature. It turns out that the formulae fit experimental facts well if the first assumption is made. It has not been derived theoretically; there is no hope of doing so until a thorough theoretical treatment of polygonization is available. Moreover, the gain of strain energy when a polygonization block breaks continuity with the parent lattice would have to be taken into account, so that even if a theoretical treatment were possible, it would probably be found at best that it was only a rough approximation.

With these assumptions we obtain the rate of nucleation in isothermal recrystallization as follows.

For the normal frequency distribution of the curvatures we write

$$n(\alpha)\delta\alpha=\frac{A}{(2\pi)^{\frac{1}{2}}\sigma}\exp\left[-\frac{1}{2}\left(\frac{\alpha-\alpha_0}{\sigma}\right)^2\right]\delta\alpha, \qquad \ldots\ldots(1)$$

where $n(\alpha)\delta\alpha$ is the number of curvatures occurring in the range α to $(\alpha+\delta\alpha)$. α_0 is the mean of the distribution, σ is the standard deviation.

I. Put $t=\tau\rho=\tau/\alpha$. Then $\delta t=-(\tau/\alpha^2)\delta\alpha$ or $\delta\alpha=-\dfrac{\alpha^2}{\tau}\,.\,\delta t=-\dfrac{\tau}{t^2}\,.\,\delta t.$

If $N(t)\,\delta t$ is the number of nuclei becoming active in the time interval t to $(t+\delta t)$, we have

$$N(t)\,\delta t=n(\alpha)\,\delta\alpha=\frac{A}{(2\pi)^{\frac{1}{2}}\sigma}\,\frac{\tau}{t^2}\exp\left[-\frac{1}{2}\left(\frac{\alpha-\alpha_0}{\sigma}\right)^2\right]\delta t \text{ (omitting the minus sign)}$$

where α to $(\alpha+\delta\alpha)$ are the curvatures of these nuclei.

Let t_{m} be the time at which the most frequently occurring nuclei (i.e. those with curvatures α_0) become active.

Then $t_{\mathrm{m}}=\tau/\alpha_0$. Putting $\sigma=\alpha_0/2$, we have

$$N(t)=\left(\frac{2}{\pi}\right)^{\frac{1}{2}}A\exp\left[-2\left(\frac{t_{\mathrm{m}}}{t}-1\right)^2\right]\frac{t_{\mathrm{m}}}{t^2}\,. \qquad \ldots\ldots(2)$$

II. Put $t=\tau\rho^2=\tau/\alpha^2$. By an exactly similar argument we derive for this case

$$N(t)=\frac{A}{(2\pi)^{\frac{1}{2}}}\exp\left[-2(t_{\mathrm{m}}^{\frac{1}{2}}-t^{\frac{1}{2}})^2/t\right]\,.\,\frac{t_{\mathrm{m}}^{\frac{1}{2}}}{t^{3/2}}\,. \qquad \ldots\ldots(3)$$

In both cases, t_m is a function of α_0, and the latter is determined by the deformation ϵ. For a given value of ϵ, t_m varies with τ, which depends on the temperature T. A is a function of ϵ only.

We can effect a further simplification by choosing our time-scale so that $t_m = 1$. We then have for the first case (with $\sigma = \alpha_0/2$)

$$\text{I.} \quad N(t) = K_1(\epsilon)\exp[-2(1-t)^2/t^2] \, . \, \frac{1}{t^2} \, . \qquad \ldots\ldots (4)$$

where $K_1(\epsilon)$ has been written in place of A.

For case II we have

$$N(t) = K_2(\epsilon)\exp[-2(t^{-\frac{1}{2}} - 1)^2] \, . \, t^{-3/2}. \qquad \ldots\ldots (5)$$

These curves were plotted. It was found that the maximum value of $N(t)$ did not occur at $t = t_m = 1$, but at $t = 0{\cdot}74$ (case I) and at $t = 0{\cdot}63$ (case II). The time-scales were again adjusted so that in each case the maximum value of $N(t)$ came at $t = 1$, and values of K were chosen to bring the value of $N(t)_{\max}$ to unity (Figure 5, curves A and B). Thus the curves are 'fitted' at two points, (0, 0) and (1, 1) and can be compared with each other and with Anderson and Mehl's experimental results, if these are fitted at the same points. This has been done in Figure 5. The fitting at (1, 1) was not exact, because the set of points marked with a square had to be extrapolated to a maximum, while in the two series indicated by circles and triangles there were so few points that the position of the maximum had to be estimated approximately. The three sets of experimental points, normalized in this way, are seen to approximate to a standard curve of a shape very similar to that of the theoretical curve A, though the latter is displaced somewhat along the time-axis relative to the experimental points. Curve B is evidently of a wrong shape. The shape of curve A is little altered by using different values of σ (curves C and D).

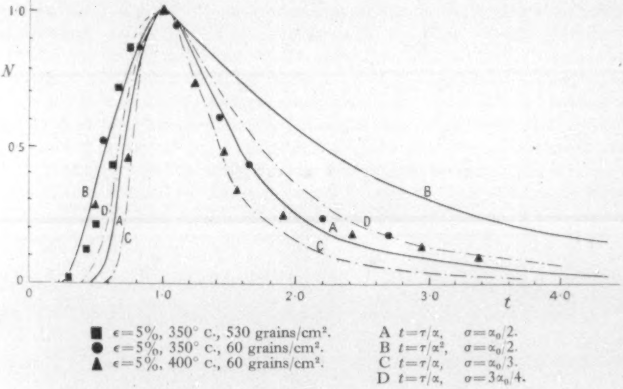

Figure 5. Normalized plot of N against t, calculated on several different assumptions. Anderson and Mehl's normalized results plotted for comparison.

Since (1, 1) is an arbitrarily chosen fitting point, any other point may be used instead of it; in other words, it is permissible to change the time-scales of the theoretical curves so as to obtain a fit elsewhere than at the maximum. The best fit is in fact obtained by compressing curve D ($\sigma = 3\alpha_0/4$) by 10% along the time-axis and this is drawn in Figure 6. It should be pointed out that the changes of scale and two-point fitting operations do not invalidate the plots as tests of the *form* of functions (2) and (3). These are functions of degree greater than two and the operations in question cannot force an artificial correspondence; they are merely necessary because of our ignorance of the values of the theoretical constants A and t_m.

The fact that hypothesis I gives a much better fit than II shows that the variation of the time required by a nucleus to become active does not depend *very* sharply on the curvature. This is in accord with experimental findings by Guinier and Tennevin (private communication) on the polygonization time of crystals stretched by different amounts.

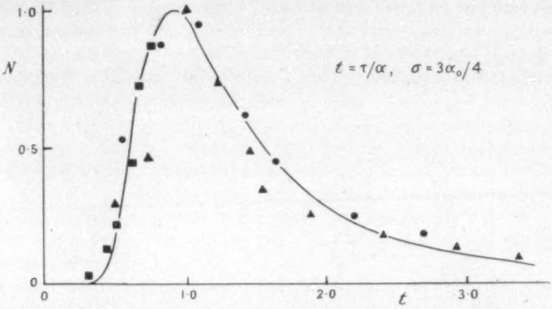

Figure 6. Normalized plot of N against t, compressed 10% along t axis.

The method of plotting used brings out the important point that the experimentally found variation of N with time follows approximately a curve of the same shape (that is, obeys a function of the same form for different temperatures and grain sizes. This is as theory predicts, for temperature has no effect on the factor K_1 in the normalized equation (4), while a change in grain size, as is argued later, would lead to a change in K_1, and also to some change in σ. The latter change has a relatively slight effect on the shape of the curve, while any change in K_1 is eliminated by the method of plotting.

§ 4. COMPARISON WITH OTHER EXPERIMENTAL RESULTS

Anderson and Mehl found that N varied with the temperature according to a relation of the form $N = N_0 e^{-B/T}$. This is to be expected on the present theory, since the same processes govern nucleation at all temperatures. The rate at which they occur is inversely proportional to τ. τ is related to the rate of diffusion of dislocations, which is governed by an activation energy. We may therefore assume that τ depends on temperature according to a law of the form $\tau = \tau_0 e^{C/T}$, where C is a constant.

Let us suppose for a moment that the values of $N(t)$ being compared for different temperatures correspond to a stage at which the same total number of nuclei has been formed. Now we can show quite easily that this is equivalent to comparing values of $N(t)$ at times t which are always the same fraction f of t_{m}.

We have by integration that the number of nuclei formed at time ft_{m} is

$$\int_0^{ft_{\mathrm{m}}} N\,dt = A\left(\frac{2}{\pi}\right)^{\frac{1}{2}} \cdot \int_0^{ft_{\mathrm{m}}} \cdot \frac{t_{\mathrm{m}}}{t^2} \exp\left[-\beta\left(\frac{t_{\mathrm{m}}}{t}-1\right)^2\right] dt,$$

where β depends on the standard deviation.

If we write $x = t_{\mathrm{m}}/t - 1$ then the number of nuclei is

$$-A\left(\frac{2}{\pi}\right)^{\frac{1}{2}} \cdot \int_{\infty}^{\frac{1-f}{f}} \exp(-\beta x^2) \, . \, dx.$$

This integral must be independent of t_{m}, since both the limits are constants; thus at times ft_{m} the number of nuclei formed, ν, is the same for different temperatures of recrystallization. Since t_{m} is proportional to τ for a given extension, $t_{\mathrm{m}} = De^{C/T}$ where D is constant. From equation (2), putting $t = ft_{\mathrm{m}}$, we find that $\log N(ft_{\mathrm{m}}) = \log E - \log t_{\mathrm{m}}$, where E is a function of f only. Therefore $N(ft_{\mathrm{m}}) = E/De^{-C/T}$, and this is in agreement with Anderson and Mehl's conclusions.

Strictly, Anderson and Mehl did not compare values of $N(t)$ at times $t = ft_{\mathrm{m}}$, but at times at which a constant fraction 0·2 of the metal had recrystallized. Since, however, the rates of nucleation and growth are found to vary at almost the same rate with temperature (i.e. the activation energies do not differ much for low degrees of deformation), it can easily be seen that almost identical results must be obtained whether N is compared (i) when the same total number of nuclei has been formed, (ii) when the same fraction of metal has recrystallized. This is certainly true over the small range of temperature, about 100° c., covered by the experiments.

The variation of N with ϵ, the degree of deformation, is too complicated a problem to be amenable to theoretical treatment. The one thing which may be said is that one would expect N to approach a limit with increasingly heavy deformation. This is because Taylor and Quinney (1934) found that there is a limiting retained energy of cold work, corresponding presumably to a limiting concentration of dislocations, and therefore of potential nuclei. One would thus expect the rate of increase of N with ϵ to fall off beyond a certain value of ϵ, until N becomes almost independent of ϵ. Anderson and Mehl's curves, which only go up to about 12% elongation, give no indication of this.

There remain now some qualitative aspects of the theory to be compared with experimental results.

(i) Weakly deformed fine-grained aluminium gave a curve of $N(t)$ against the time which barely passes the point of inflection, while heavily deformed fine-grained metal (to quote Anderson and Mehl) "either shows a maximum in N at short times or steadily decreasing values of N from the beginning". This can be understood when it is remembered that N can be determined only up to the time when most of the sample has recrystallized. Anderson and Mehl report that the rate of growth G increases less rapidly with degree of deformation than does N. At high degrees of deformation, quite a late stage of the 'master curve' of N against t may be reached before all the deformed matrix is swallowed up by the new grains, while with weak deformation, very few nuclei are formed and these grow relatively quickly, so that the whole sample has recrystallized before the maximum is reached. The fact that maxima occur at short times in the first case is simply due to the sharper curvatures present in heavily deformed metal. Since determinations of N could, by the nature of the experimental method, only be made periodically, it may be that sometimes the rising part of the curve was missed altogether in heavily deformed specimens and N appeared to decrease with time from the start.

(ii) The profound effect of grain-size on the magnitude and time-dependence of N must be explained. Fine-grained and coarse-grained metal differ essentially because in the former case the total area of grain boundary is greater. In Anderson and Mehl's work, comparison was made between specimens having respectively 530 and 60 grains per cm^2 of cross section. The former has about three times the grain boundary area of the other; this shows that the effect of this area is a powerful one. The retardation of nucleation is carried to the extreme when single crystals are recrystallized. Thus a single crystal of aluminium, pulled by say 15%, may not recrystallize after an hour at 450° C., while the induction time for similarly treated polycrystalline metal is only a few seconds.

It has been known for many years that the state of deformation near the grain boundaries differs radically from that within the grain; more recently it has become clear that the difference lies primarily in the great inhomogeneity of slip near the boundaries. Boas and Hargreaves (1948) have made the most thorough study of this point, by actually tracing the variation, on a microscopic scale, of the deformation across the grains of aluminium specimens. They found that the deformation was more or less uniform in the body of each grain but changed sharply in the boundary regions, increasing or decreasing according as the adjacent crystal was more or less strongly deformed. Greenough (1947) was able to confirm, by means of X-rays, the great variation in internal strain between different grains in deformed aggregates; this variation is due to the different amounts of slip in the grains. There is then a much greater shear gradient near the boundaries than inside the grains, and this means, as we have seen earlier, that the nuclei are more sharply curved—that is, have a greater value of α_0. Nucleation would therefore be expected to be largely concentrated at the boundaries, and this is confirmed by experiment (van Arkel 1932, Polanyi and Schmid 1924).

The distribution of values of α must, then, refer to the boundary regions rather than to the inside of the grains; the latter only furnish a relatively small 'tail' to the distribution. Further, for a given extension the total number of nuclei will increase as the grain size decreases, that is, as the total area of boundary increases. Since the inhomogeneity of slip will be the more marked the smaller the grain, the mean sharpness of curvatures α_0 will also increase as the grain size decreases. The time at which $N(t)$ is a maximum depends only upon α_0, and not upon the total number of potential nuclei. Thus by studying the dependence of this time, and of the absolute values of N, on the grain size, further evidence of the state of deformation at the grain boundary could perhaps be obtained.

In a single crystal, deformed *homogeneously*, no shear gradient can arise, and recrystallization is very much slower than with fine-grained metal. Apparently homogeneous deformation is not usually truly so. Thus it has usually been considered that a long crystal extended in tension undergoes homogeneous slip in the parts remote from the grips. Often such crystals are full of deformation bands occasioned by uneven yielding of different parts of the crystals, and recrystallize readily; but in other cases, particularly with crystals of hexagonal symmetry, extension does seem to be very nearly homogeneous, and then recrystallization does not occur, even at the highest temperatures. In such cases the local curvatures are evidently so weak that they cannot function as nuclei. Thus Honeycombe (private communication) stretched cadmium crystals by as much as 200% without getting them to recrystallize on subsequent annealing; Maddin (private communication) using somewhat weaker deformations with α-brass, found the same.

(iii) There is a statistical connection between deformation and recrystallization textures. This has already been mentioned in §2 in connection with the work of Burgers and Louwerse. The local curvatures should be related to the main body of the crystal by a rotation about an axis approximately in the glide-plane and perpendicular to the glide-direction, and on this basis it is possible to account for the sharp recrystallization textures found with heavily compressed crystals.

With a weakly deformed crystal, the preferential orientation of the new grains is less well marked. Burgers and Basart (1928) concluded, from their experiments, that the connection was merely a weak statistical one, without any well-marked axes of rotation connecting the orientations of the new grains with that of the parent crystal. Some unpublished experiments by the writer suggest that this apparently almost haphazard orientation is probably due to the incidence of 'pencil glide' in aluminium (in common with many other substances). The glide-direction [110] is fixed, but the glide-plane can be any plane containing this direction, although a particular {111} plane is preferred. The cross-slip on other planes is particularly marked in fairly weakly deformed crystals, and it follows that the local curvatures which turn into nuclei on annealing are rotated with respect to the parent crystal about several of the axes. An experimental redetermination of the recrystallization textures of such crystals suggested, on analysis, that this is the explanation of the low correlation of orientation. The same study showed (though more experiments will be needed to make the point certain) that the first recrystallization grains to form have on the average an orientation further removed from that of the parent crystal than the later ones. This is an excellent confirmation of the idea that local curvatures turn into nuclei on annealing because the more sharply bent ones (which become active first) will usually contain lattice elements more remote in orientation from the parent lattice than the weakly bent ones.*

(iv) As was mentioned in §1, a low-temperature anneal sometimes affects subsequent recrystallization behaviour. Kornfeld and his co-workers showed that the number of recrystallization grains was almost invariably increased, on the average by about 50%, by a prior recovery treatment in monocrystalline aluminium, while in polycrystalline specimens the number of grains fell, statistically, by half. In view of evidence, advanced in one of the papers, that the

* *Note added in proof.* Recent work has shown that selective *growth* of nuclei of particular orientations relative to the parent crystal is an important factor in leading to recrystallization textures. This has been shown in several laboratories to be true for secondary recrystallization, but recently corresponding results have been obtained respecting primary recrystallization (Kronberg and Wilson 1949, Beck, Sperry and Hsun Hu, unpublished). This factor can only operate effectively if the number of potential nuclei is very large and if they have all possible orientations; otherwise growth must necessarily occur from the available nuclei, whatever their orientations. In view of these new findings it is felt that it is no longer correct to invoke oriented nucleation as being *essential* to explain recrystallization textures; this is not to say, however, that it plays no part.

rate of growth is unaffected by recovery, the authors argue from their result that the mean value of N is increased by recovery in monocrystalline metal and decreased in the polycrystalline. Anderson and Mehl quote other references mentioning a retardation of subsequent recrystallization, but in their own experiments no such effect appears.

The present theory as it stands requires that a recovery anneal should always accelerate subsequent nucleation, because such an anneal brings about the initial stages of polygonization in the local curvatures. As a consequence of this the plot of $N(t)$ against t is everywhere shifted to the left; therefore every potential nucleus becomes active a little earlier than it would have done in the absence of the recovery anneal and, since the rate of growth is unaffected, more grains will be formed before the parent crystal is consumed. Unless the recovery anneal is carried out at a temperature relatively close to the recrystallization temperature in question, the effect should be slight: it will be the more difficult to discern because the nucleation rate in deformed *single crystals* is liable to great statistical fluctuation.

The explanation of the opposite results with polycrystalline aluminium may be in terms of a reduction of internal stress in the grain boundary regions by the recovery treatment. It has already been pointed out that most of the potential nuclei are in these regions; these curvatures are as it were 'held in position' by the internal stresses at the boundary, and if these stresses can be partly relieved, many of the captive dislocations will be able to complete their journeys to the boundary and thereby decrease the concentration of nuclei. Such localized plastic flow would be associated with viscous actual displacement of neighbouring grains at their boundaries; such has been shown to occur in aluminium polycrystals by Hanson and Wheeler (1931) and other authors. The existence of this type of flow has been demonstrated particularly clearly with *bicrystals* of tin (King, Cahn and Chalmers 1948) and aluminium (Cahn, unpublished work). In these experiments it was found that the flow does not remain viscous but soon slows down, and the displacement reaches a limiting value depending on temperature and stress. Similarly, the physical properties of cold-worked metals affected by a recovery anneal also reach limiting values different from those obtaining in recrystallized metal. Moreover, Kornfeld and Pavlov (1934) found that when the number of recrystallization grains was plotted against the time of prior recovery anneal, the former quantity reached a limit beyond which it did not decrease further; if there is a decrease in the number of local curvatures down to a certain limit, occasioned in turn by limited viscous flow at the boundaries, this observation can be understood. If annealing had been long continued or carried out at a high temperature one would have expected an increase once again in the number of grains, on subsequent recrystallization, because of the progressive polygonization of the curvatures. In a plot of annealing temperature against the number of grains, with a constant annealing period, the number in fact begins to rise again for a recovery temperature near to the recrystallization temperature used. The temperature dependences of polygonization rate and of boundary viscosity will be different since they are unrelated quantities. It may be that the discrepancies between the results of different workers on the effect of a recovery treatment on the recrystallization of polycrystals are due to the operation of these two opposing processes, the preponderance of one or the other or their balancing out, depending on the temperature and material used.

Single crystals cannot, of course, recover by boundary flow, and in their case recovery of physical properties must be due entirely to some different mechanism, such as the mutual annihilation of dislocations (Burgers 1947a, b).

§ 5. COMPARISON WITH OTHER THEORIES

It is not possible to make a detailed comparison, because the theories advanced have often been couched in vague terms. Though numerous, they fall into two main groups, differing otherwise only in minor aspects. One school holds that nucleation takes place in the most heavily distorted regions of the lattice, the other that it occurs by the growth of spots in the lattice which are relatively little deformed (so-called preformed nuclei). Burgers (1941) marshalled the evidence for both these views and held that a firm decision between them is not possible. Most of the points which he regarded as evidence for the second view have been shown in the present paper to follow also from the first. It has always been assumed that if nucleation takes place in heavily distorted regions, the nuclei must be quite unrelated in orientation to the mother crystal; and the fact that this is contrary to experiment is held as the main point in favour of the second school. The present theory does not involve this assumption and reconciles the existence of preferred orientation with the experimentally observed occurrence of nuclei in the most heavily distorted regions (but see footnote on p. 333).

The adherents of the first school have in part developed theories based on critical sizes of nuclei. The fullest treatment is that due to Becker and Döring (1935).* In a recent paper, Turnbull (1948) seeks to reconcile this theory with the existence of an induction period (one of its chief stumbling-blocks). This period is held to be due to the time needed for a minimum sized nucleus to build up by diffusion of atoms, and in this respect is *formally* similar to the present theory according to which diffusion of dislocations is the basic process. The arguments advanced to explain preferred orientation seem less sound, however, and it seems unlikely that nucleation in the sense of Becker and Döring could lead to this phenomenon.

Another recent theory is that due to Petersen (1947), who also considers atomic diffusion to be the key to the problem. Such diffusion is held to lead to a bodily rotation of crystal fragments during annealing, resulting in a decrease in elastic energy; when the process has gone far enough the free energy of the system can be further reduced if the nucleus begins to grow at the expense of its surroundings. This theory has therefore the advantage over the present one that it considers in detail the exact conditions under which a potential nucleus becomes active and is able to grow. However, it does not draw into consideration the part played by dislocations in setting up points of internal stress and in relieving this again on annealing, a part which, as the existence of the phenomenon of polygonization shows, must be an essential one The same criticism applies to Turnbull's theory.

It should be mentioned that the idea that the motion of dislocations is at the root of recovery and recrystallization has been suggested previously by Dehlinger (1941) and more recently by Burgers (1947 a). Burgers (1947 b) has also sought to account for the increase of $N(t)$ with time by invoking stimulated crystals, which can only begin to grow when other growing grains approach their points of nucleation. It appears, however, that a special and exact orientation relationship between stimulating and stimulated crystal must exist and, for this reason alone, it hardly seems likely that the very considerable acceleration of $N(t)$ after the induction period can be explained on this basis.*

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* It should be pointed out that in theories of this type, every point of potential nucleation has a certain *probability* of turning into an active nucleus, while in the present theory each such point will do so after a *definite time*.

ACKNOWLEDGMENTS

The author gladly acknowledges his debt to Professor E. N. da C. Andrade, Dr. H. M. Finniston, Professor N. F. Mott and Dr. E. Orowan, who read the manuscript and pointed out various errors and obscurities. Thanks are also due to the Chief Scientist, Ministry of Supply, and to the Director A.E.R.E. for permission to publish.

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* *Note added in proof.* P. A. Beck (1949) has independently suggested that recrystallization nuclei are formed by the polygonization of local high-stress regions.

Nuclear Induction Signals for Long Relaxation Times

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Communicated by R. E. Peierls; MS. received 29th August 1949

ABSTRACT. Equations of motion, previously derived by Bloch (1946), for the nuclear magnetic moment in a nuclear induction experiment are applied to the steady state reached when the strong magnetic field H_z is swept sinusoidally about its resonance value. Different approximations for the shapes and sizes of nuclear induction signals are derived for various limiting cases, depending (*a*) whether the passage of H_z is ' adiabatic ' or ' extremely non-adiabatic ' (very weak radio-frequency field) and (*b*) whether the relaxation times are shorter or longer than the period of the sweep of H_z. It is found that quite large signals may be induced by even a very weak radio-frequency field if the relaxation times are very long.

§1. INTRODUCTION

In a paper, hereafter referred to as A, Bloch (1946) has derived the equations of motion for the total nuclear magnetic moment in a nuclear induction experiment. In this paper we shall assume an experimental set-up in which the substance to be investigated is acted upon by a strong magnetic field H_z in the z-direction and by a weak magnetic field of constant magnitude H_1, the direction of which rotates about the z-axis with constant angular frequency p. We assume H_z to be equal to its resonance value H_0 plus a field varying sinusoidally with time with period τ_S and of amplitude H_S, large compared with H_1 but small compared with H_0. In this case the equations of motion take the form of a set of simultaneous differential equations with variable coefficients and depending on five parameters. These parameters are the Larmor period of precession τ_L, the sweep-period τ_S of the field H_z, the 'resonance time' τ_R, and the two relaxation times τ_1 and τ_2. An exact solution of these equations for any values of these five parameters seems impracticable, but for a number of cases in which the parameters satisfy certain inequalities approximate solutions can be found.

In reference A the limiting case of 'adiabatic passage' is discussed, i.e. the case of the resonance time τ_R very long compared with the Larmor period τ_L. In a paper, hereafter referred to as B, Jacobsohn and Wangsness (1948) treat the opposite limiting case, which we shall denote as 'extreme non-adiabatic passage', where τ_R is very small compared with τ_L. The results of both A and B are valid only if both the relaxation times τ_1 and τ_2 are small compared with the sweep period τ_S. It was kindly pointed out to the author by Dr. J. S. Gooden that both the shapes and maximum values of nuclear induction signals are radically different from those discussed in A and B if one or both of τ_1 and τ_2 are larger than the sweep period.

In connection with the use of nuclear induction experiments for measuring rapidly varying magnetic fields, and with other applications, experimental set-ups using very short sweep periods are of some interest. The main aim of this paper is to discuss the steady state reached with a sinusoidal sweep of period small compared with τ_1 (and possibly τ_2). Formulae for the components of the induced nuclear magnetic moment in such cases are derived in § 4. To facilitate comparison with these cases, and since the results of A and B do not strictly apply to sinusoidal sweep, equivalent formulae are also derived in § 3 which are valid when the sweep period is larger than both τ_1 and τ_2. The results obtained for the amplitudes of the nuclear induction signals in these various cases are discussed in § 5. A glossary of symbols used is appended.

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§ 2. NOTATION AND ASSUMPTIONS

We take as the starting point of the present paper the equations (42) of A, derived by means of classical arguments, without essential alterations. For convenience we shall adopt a somewhat different notation. We take over the definitions of section 4 of reference A for the two relaxation times. We call the 'longitudinal' relaxation time, connected with the establishment of thermal equilibrium, τ_1 and the 'transverse' relaxation time, connected with processes not affecting the nuclear spin energy, τ_2. We call the amplitude and angular frequency of the rotating field, H_1 and p respectively; the amplitude and repetition period of the sweep of the field H_z in the z-direction H_S and τ_S respectively. The resonance value H_0 for the field in the z-direction is equal to p/γ and the period τ_L of the Larmor precession is given by

$$\tau_L = 2\pi/\gamma H_0. \qquad \ldots\ldots (1)$$

γ is the gyromagnetic ratio of the substance considered and for simplicity we assume it to be positive. We define the 'resonance time' τ_R as the time taken for H_z to pass through the 'resonance band' between $(H_0 - \frac{1}{2}H_1)$ and $(H_0 + \frac{1}{2}H_1)$. τ_R is approximately equal to $(\tau_S H_1/2\pi H_S)$.

The form of the various results to be discussed depends only on the ratios of the five parameters mentioned above, each of the dimension of time, to each other. We shall therefore find it convenient to express all quantities of the dimension of time as multiples of $\tau_L/2\pi$, i.e. we define the following dimensionless quantities:

$$\left.\begin{aligned} t &= (\text{time in seconds})\,(\gamma H_0), \\ T_S &= \tau_S(\gamma H_0);\ \ \omega = 2\pi/\tau_S, \\ T_R &= \tau_R(\gamma H_0) = H_1/\omega H_S, \\ T_1,\ T_2 &= \tau_1(\gamma H_0),\ \tau_2(\gamma H_0). \end{aligned}\right\}$$

Where no ambiguity exists we shall refer to t, T_1, T_2, T_R and T_S by the names of the equivalent times. We also define the ratio $k = H_S/H_1 = \tau_S/2\pi\tau_R$.

We shall assume throughout that the sweep amplitude of H_z is small compared with its resonance value but large compared with the amplitude of the rotating field, i.e.

$$1 \ll k \ll H_0/H_1. \qquad \ldots\ldots (2)$$

We also restrict ourselves to sinusoidal sweep about the resonance value H_0 as mean, i.e. we take H_z as the following function of time

$$H_z(t) = H_0 + kH_1 \sin \omega t. \qquad \ldots\ldots (3)$$

It follows from the inequality (2) that the resultant magnetic field at any time does not differ appreciably from a field H_0 in the z-direction. The thermal equilibrium value for the resultant nuclear magnetic moment per unit volume at any time can then be replaced by M_0, its value for a field H_0. We resolve the actual resultant nuclear magnetic moment per unit volume into $m(t)M_0$, the component in the z-direction, and into two components rotating about the z-axis with angular frequency $p: u(t)M_0$ in phase with and $v(t)M_0$ in quadrature with the rotating field H_1. The three functions $m(t)$, $u(t)$ and $v(t)$ are then dimensionless.

Finally we combine the equations for $u(t)$ and $v(t)$ into a single equation involving the complex function

$$F(t) = v(t) + i\,u(t)$$

and the complete set of equations determining m, u and v is then

$$\frac{dF(t)}{dt} + \left[\frac{1}{T_2} + ik\sin\omega t\right]F(t) = -m(t), \qquad \ldots\ldots (4)$$

$$\frac{dm(t)}{dt} + \frac{m(t)-1}{T_1} = v(t). \qquad \ldots\ldots (5)$$

The general solution of the inhomogeneous differential equations (4) and (5) consists of a truly periodic 'steady state' solution plus any multiple of the solutions of the homogeneous equations obtained by omitting the inhomogeneous term $(-1/T_1)$ in (5). Because of the damping terms (F/T_2) and (m/T_1) in (4) and (5) respectively, the solutions of the homogeneous equations decay with time and only represent 'transients'. Throughout this paper we shall only consider the truly periodic 'steady state' solutions of (4) and (5), the period of which is equal to the sweep period $2\pi/\omega$. Now $\sin\omega t$ is an 'odd' function in the sense that $\sin\omega t = -\sin[\omega(t+\pi/\omega)]$. Hence, as pointed out in B, it follows from (4) and (5) that $u(t)$ is an 'odd' function, $v(t)$ and $m(t)$ are 'even' functions, i.e.

$$\left.\begin{aligned} m(t+\pi/\omega) &= m(t), \\ v(t+\pi/\omega) + iu(t+\pi/\omega) &= v(t) - iu(t). \end{aligned}\right\} \qquad \ldots\ldots (6)$$

We shall derive all formulae for a single sweep only (half-cycle) for $-\pi/2\omega < t < \pi/2\omega$. The values of u, v and m for all other times are then given by the periodicity conditions (6). There is, however, not necessarily any symmetry between the parts before and after resonance for any single sweep, i.e. between $F(t)$ and $F(-t)$ etc.

For some cases we shall find it useful to solve, instead of the pair of differential equations (4) and (5), a pair of integral equations, equivalent to (4) and (5) for periodic solutions, i.e.

$$F(t) = -\exp\{i(k/\omega)\cos\omega t\}\int_{-\infty}^{0} d\tau\, m(t+\tau)$$
$$\times\exp\{(\tau/T_2) - i(k/\omega)\cos\omega(t+\tau)\}, \qquad \ldots\ldots (7)$$

$$m(t) = 1 + \int_{-\infty}^{0} d\tau\, v(t+\tau)\exp(\tau/T_1). \qquad \ldots\ldots (8)$$

A qualitative picture in terms of the following three competing processes may be of help in visualizing the physical significance of the equations (4) and (5) or (7) and (8): (i) For any value of $H_z(t)$ the resultant nuclear magnetic moment tends to take up a position parallel or antiparallel to that of an 'effective field' composed of a field $(H_z(t)-H_0)$ in the z-direction plus the rotating field H_1. (ii) The resultant moment cannot change appreciably in a time small compared with the Larmor period and will therefore lag behind the 'effective field'. (iii) Due to thermal motion, etc., the resultant moment tends to return to its thermal equilibrium value of unity for $m(t)$ and of zero for $u(t)$ and $v(t)$ in times of the order of magnitude of T_1. In addition inhomogeneities in the magnetic field throughout the substance considered (due to interatomic fields or irregularities in the external fields) tend to reduce the total values of $u(t)$ and $v(t)$ to zero without appreciably affecting $m(t)$. Thus $u(t)$ and $v(t)$ tend to return to their equilibrium value zero in times of the order of magnitude of T_2, where

$$T_2 \leqslant T_1. \qquad \ldots\ldots(9)$$

§ 3. RELAXATION TIMES SHORTER THAN THE SWEEP PERIOD

We briefly review in this section some of the results for the case where both relaxation times are much smaller than the sweep period,

$$T_1,\ T_2 \ll 2\pi/\omega. \qquad \ldots\ldots(10)$$

If (10) holds then m, u and v differ from their equilibrium values only for a small fraction of each sweep, near the passage of H_z through resonance, except for one special case which we discuss below. Over this small fraction the sweep is approximately linear, i.e. we can replace $\sin\omega t$ by ωt and $\cos\omega t$ by $(1-\frac{1}{2}\omega^2t^2)$ in equations (4), (5), (7) and (8).

A. *Adiabatic Passage.*

The criterion for passage being 'adiabatic' is that the resonance time T_R is much larger than the Larmor period, i.e.

$$1 \gg 2\pi\omega k = (2\pi)^2(H_S/\gamma H_0 H_1 \tau_S). \qquad \ldots\ldots(11)$$

In reference A approximations are given valid for the two limiting cases of T_R much smaller and much larger respectively than both relaxation times. We consider here only the first case, i.e.

$$2\pi \ll (k\omega)^{-1} \ll T_1,\ T_2 \ll 2\pi/\omega. \qquad \ldots\ldots(12)$$

The qualitative behaviour of the resultant nuclear magnetic moment is then as follows. At the beginning of a sweep $m(t)$ and $F(t)$ remain approximately at their equilibrium values unity and zero respectively until $H_z(t)$ approaches its resonance value (reached at $t=0$). The direction of the 'effective field' then changes and reverses its sign in a time of the order of magnitude of T_R. This variation is slow enough for the nuclear moment to follow it without appreciable time-lag (i.e. $v \ll u$); at $t=0$ $u(t)$ is approximately unity and $m(t)$ minus unity. During a time of the order of magnitude T_1 after resonance $m(t)$ changes from minus unity to its equilibrium value plus unity and remains constant until $H_z(t)$ again approaches resonance on the backward sweep. During the backward sweep the pattern is the same as on the forward sweep except that u has the opposite sign. Note that the resultant nuclear moment follows in alternate sweeps directions respectively equal to and opposite to the 'effective field', since $m(t)$ is an 'even function' and $u(t)$ 'odd', whereas $(H_z(t)-H_0)$ is 'odd' and the constant

H_1 'even'. The approximate expressions, derived from (4) and (5), for $-\pi/2\omega<t<\pi/2\omega$ are

$$m(t)=-k\omega t(1+k^2\omega^2t^2)^{-1/2}+\eta(t), \qquad \ldots\ldots(13)$$

$$u(t)=-(1+k^2\omega^2t^2)^{-1/2}, \qquad \ldots\ldots(14)$$

$$v(t)=-k\omega(1+k^2\omega^2t^2)^{-3/2}, \qquad \ldots\ldots(15)$$

where

$$\left.\begin{aligned}\eta(t)&=0 && \text{for } t<0\\ &=2[1-\exp(-t/T_1)] && \text{for } t>0.\end{aligned}\right\} \qquad \ldots\ldots(16)$$

A rough sketch of these expressions is given in Figure 1.

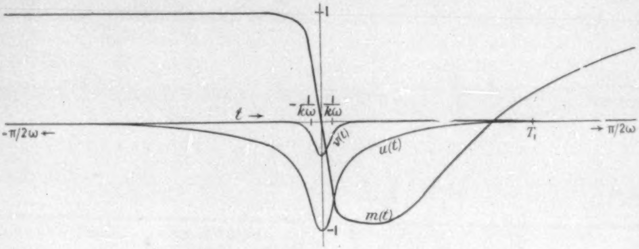

Figure 1. $2\pi \ll (\omega k)^{-1} \ll T_1,\ T_2 \ll 2\pi/\omega$.

B. *Extreme Non-adiabatic Passage.*

We consider here the opposite limiting case where the resonance time T_R is much shorter than the Larmor period, i.e.

$$1 \ll 2\pi\omega k=(2\pi)^2(H_S/\gamma H_0 H_1 \tau_S). \qquad \ldots\ldots(17)$$

This condition implies that the rotating field H_1 is 'weak'. In this case the 'effective field' changes its direction during the passage of $H_z(t)$ through resonance in a small fraction of a Larmor period and the nuclear magnetic moment cannot 'follow the field' rapidly enough. Consequently $m(t)$ will never vary very much from its equilibrium value plus unity; both $u(t)$ and $v(t)$ will have maximum values small compared with unity but will not return to their equilibrium values, zero, immediately after passage through resonance.

Thus, if (17) holds, we can replace $m(t+\tau)$ in (7) by unity to get an approximate but explicit expression for $F(t)$. We shall not restrict ourselves in this section to times t small compared to the sweep period but the main contribution to the integral (7) comes from values of the integrand such that $\omega\tau \lesssim \omega T_2 \ll 2\pi$. This enables us to derive the following approximation for (7)

$$F(t)=-\int_{-\infty}^{0} d\tau \exp\left\{\left[\frac{1}{T_2}+ik\sin\omega t\right]\tau+\frac{ik\omega\tau^2}{2}\cos\omega t\right\}. \qquad \ldots\ldots(18)$$

The form of the integral (18) depends on the value of the dimensionless parameter $(k\omega T_2^2)$ and was discussed for general values of it in reference B but assuming a linear instead of sinusoidal sweep. We shall merely discuss the two limiting cases of $k\omega T_2^2$ very small and very large respectively compared with unity, but for sinusoidal sweep.

(i) $k\omega T_2^2 \ll 1$.

In this case the term involving $k\omega\tau^2$ in (18) can be omitted altogether and we obtain the following approximation:

$$u(t) = T_2\{1+(kT_2\sin\omega t)^2\}^{-1}(kT_2\sin\omega t), \qquad \ldots\ldots(19)$$

$$v(t) = -T_2\{1+(kT_2\sin\omega t)^2\}^{-1}. \qquad \ldots\ldots(20)$$

Both $u(t)$ and $v(t)$ are appreciably less than their maximum value at the end of a sweep only if kT_2 is large compared with unity. Sketches are given in Figures 2 and 3 for two values of kT_2.

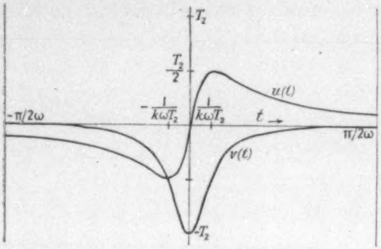

Figure 2. $2\pi\omega k \gg 1$; $\omega k T_2^2 \ll 1$; $kT_2 = 5$.

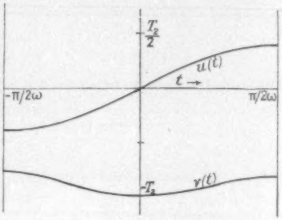

Figure 3. $2\pi\omega k \gg 1$; $\omega k T_2^2 \ll 1$; $kT_2 = \frac{1}{2}$

(ii) $k\omega T_2^2 \gg 1$.

In this case both u and v build up to their maximum values in a time of the order of magnitude of $(k\omega)^{-1/2}$ during the passage through resonance and then perform about $k\omega T_2^2$ damped oscillations which die out in a time of the order of magnitude of T_2. The term in $1/T_2$ in (18) is slowly varying compared to other terms, but $F(t)$ is appreciable over a period of about T_2 only and we can again replace $\sin\omega t$ by ωt and $\cos\omega t$ by $(1-\frac{1}{2}\omega^2t^2)$. The approximation to (18) thus obtained is

$$F(t) = \left.\begin{array}{ll} -\sqrt{\dfrac{2}{k\omega}}\exp\left[-\dfrac{ik\omega t^2}{2}\right]\left\{\Phi(\infty)-\Phi\left(\sqrt{\dfrac{k\omega t^2}{2}}\right)\right\} & \text{for } t<0, \\ -\sqrt{\dfrac{2}{k\omega}}\exp\left[-\dfrac{ik\omega t^2}{2}-\dfrac{t}{T_2}\right]\left\{\Phi(\infty)+\Phi\left(\sqrt{\dfrac{k\omega t^2}{2}}\right)\right\} & \text{for } t>0, \end{array}\right\} \qquad \ldots\ldots(21)$$

where

$$\Phi(x) = \int_0^x \exp(iy^2)\,dy = \sqrt{\frac{\pi}{2}}\left\{\mathrm{C}\left(\sqrt{\frac{2}{\pi}}x\right)+i\mathrm{S}\left(\sqrt{\frac{2}{\pi}}x\right)\right\}.$$

Each of the Fresnel Integrals $\mathrm{C}(x)$ and $\mathrm{S}(x)$, tabulated in Jahnke and Emde (1945), approaches the constant value $\frac{1}{2}$ for $x \gg 1$. A rough sketch of $u(t)$ and $v(t)$ for a representative value of $k\omega T_2^2$ is given in Figure 4.

It will be seen from the above formulae that the maximum values both for $u(t)$ and $v(t)$ are of the order of magnitude of $(k\omega)^{-1/2}$, which is much smaller than unity, for $k\omega T_2^2 > 1$ and are even smaller for $k\omega T_2^2 < 1$. It can also be shown by using (8) that, for any value of $k\omega T_2^2$, the maximum deviation of $m(t)$ from plus unity is at most $(k\omega)^{-1}$.

§4. RELAXATION TIMES LONGER THAN THE SWEEP PERIOD

All the formulae derived in § 3 hold only when both relaxation times are much smaller than the sweep period. It can be seen that the formulae there derived for the resultant nuclear magnetic moment as a function of time, equations (13)–(16) and (19)–(21), depend on the ratio of the Larmor period (2π) to T_1, to T_2 and to the resonance time T_R. But these formulae do *not* depend on the actual value of the sweep period $2\pi/\omega$ (except that the patterns repeat themselves with this period). This is due to the fact that, under the above circumstances, $m(t)$ and $F(t)$ differ from their thermal equilibrium values only for a short fraction of each sweep period (near resonance), during which time the sweep is almost linear, and conditions do not depend on the time that has elapsed since the last passage through resonance. This is no longer the case when one or both of the relaxation times are larger than the sweep period.

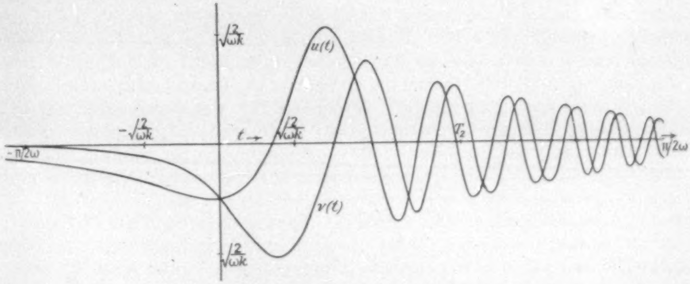

Figure 4. $2\pi\omega k \gg 1$; $\omega k T_2^2 = 20$.

We shall consider in this section the limiting case of T_1 being very large compared with the sweep period

$$T_1 \gg 2\pi/\omega. \qquad \ldots\ldots (22)$$

We still assume (2) to hold, however. Since T_2 may be much smaller than T_1 (but no larger) we shall discuss both the cases of T_2 being smaller or larger than the sweep period.

Let us imagine the rotating field H_1 suddenly switched on at a time long after the strong field H_z has been set up, so that the nuclear magnetic moment at that time has assumed its thermal equilibrium value, $m(t) = 1$ and $F(t) = 0$. During the first sweep of $H_z(t)$ through resonance after switching on H_1 the nuclear moment will have altered and, if (22) holds, $m(t)$ and possibly also $F(t)$, will not have returned to its equilibrium value when the next sweep begins; the value of $m(t)$ will be different again at the end of this sweep, and so on. Thus $m(t)$ and $F(t)$ will not be truly periodic functions with period $2\pi/\omega$ for a time of the order of magnitude T_1. These transients correspond to damped homogeneous solutions of (3) and (4) added to the truly periodic inhomogeneous solutions. In this paper we only derive formulae for the truly periodic solutions which apply to the steady state reached a time much longer than T_1 after switching on both the fields H_1 and H_z.

A. *Adiabatic Passage.*

We again treat the case of the Larmor period being much smaller than the resonance time T_R, but with T_1 satisfying (22). Formulae can be worked out for any value of T_2 but for simplicity we treat only the case where T_2 is much larger than T_R, i.e.

$$2\pi \ll T_R \ll T_2 \leqslant T_1 \gg 2\pi/\omega. \qquad \ldots\ldots (23)$$

Consider again the rotating field H_1 switched on a long time after the establishment of the strong field H_z. The passage through resonance is slow enough for the direction of the resultant nuclear moment to 'follow the effective field' or a direction opposite to it. Thus during the first sweep $m(t)$ changes from unity to minus unity during the passage through resonance, for the remaining part of this sweep $m(t)$ remains negative but decreases slightly in magnitude, due to the thermal agitation tending to restore $m(t)$ to its equilibrium value plus unity. During the next sweep (in the opposite direction) $m(t)$ returns to a positive value less than unity, during the following sweep it becomes again negative but of even smaller magnitude and so on. Finally, after a time longer than T_1, the following steady state will have been set up: at the beginning of each sweep, in either direction, $m(t)$ is zero and during the first half of the sweep builds up to a positive value C much smaller than unity. Because of (23) this exponential build up is approximately linear during the time $\pi/2\omega$. During the passage through resonance $m(t)$ changes to a negative value of the same magnitude, the absolute magnitude of the resultant of $m(t)$ and $u(t)$ remains approximately constant during the resonance time T_R, the ratio of $u(t)$ to $m(t)$ is approximately equal to that of H_1 to $(H_z(t)-H_0)$ and $v(t)$ remains much smaller than $u(t)$. During the second half of the sweep, after resonance, the algebraic value of $m(t)$ increases and $m(t)$ equals zero again when the next sweep in the opposite direction begins, builds up to the positive value C again during the first half of this sweep, changes sign again during the passage through resonance, and so on. Thus for the steady state the periodicity conditions (6) are again fulfilled.

We therefore try for an approximate solution of (4) and (5) in the following form (again for $|t| \leqslant \pi/2\omega$)

$$m(t) = -Ck\omega t(1+k^2\omega^2t^2)^{-1/2} + C\,2t\omega/\pi, \qquad \ldots\ldots (24)$$

$$u(t) = -C(1+k^2\omega^2t^2)^{-1/2}, \qquad \ldots\ldots (25)$$

$$v(t) = -Ck\omega(1+k^2\omega^2t^2)^{-3/2}. \qquad \ldots\ldots (26)$$

Substitution of these expressions into (5) for $|t| \gg T_R$ gives the following approximation to C (valid to first order in an expansion in terms of $2\pi/\omega T_1$):

$$C = \pi/2\omega T_1 \ll 1. \qquad \ldots\ldots (27)$$

These expressions hold as long as (23) is satisfied irrespective of whether T_2 is larger than the sweep period or not. This is to be expected since $F(t)$ very nearly returns to its equilibrium value zero soon after resonance during each sweep in merely 'following the effective field'. A sketch of $m(t)$, $u(t)$ and $v(t)$ is given in Figure 5.

B. *Non-adiabatic Passage.*

We again discuss here the case where H_1 is sufficiently small for the inequality (17) to be satisfied, but T_1 (and possibly T_2) is much larger than the sweep period. Consider again the rotating field H_1 suddenly switched on long after thermal equilibrium has been established. The inequality (17) ensures that during each sweep H_1 acts only as a small perturbation and changes $m(t)$ from its equilibrium value unity only by a small amount. Thus after the first passage through resonance $m(t)$ will be only slightly less than plus unity but, T_1 being very large, will not have returned to exactly plus unity after the end of the first sweep. During the second sweep $m(t)$ starts off with a value slightly less than unity and is decreased again by another small amount and so on. A time longer than T_1 after establishing the field H_1, a steady state will be reached, for which the periodicity condition (6) is satisfied exactly, when the tendency for $m(t)$ to decrease slightly during resonance is balanced by the tendency of the thermal agitation to restore $m(t)$ to its equilibrium value plus unity. In this steady state the average value of $m(t)$, which we shall denote by C, will be positive but less than unity—the smaller, in fact, the larger T_1 is. During the first sweep after switching on H_1 the maximum value of $F(t)$ will be much less than unity. If T_2 is smaller than the sweep period then $F(t)$ will have returned to zero by the beginning of the next sweep, its next maximum value is again small, and so on. In this case the maximum value of $F(t)$ after the steady state has been established will in fact be small compared to C. But if T_2 as well as T_1 is large compared with the sweep period the situation is quite different. During each sweep, after H_1 is switched on, $F(t)$ increases only by a small amount but in this case it does not return to zero before the next sweep begins. Thus after the steady state has been established the maximum value of $F(t)$ may be larger than C. This maximum value is the larger the larger T_2 is and in certain cases, as discussed below, may be of the order of unity.

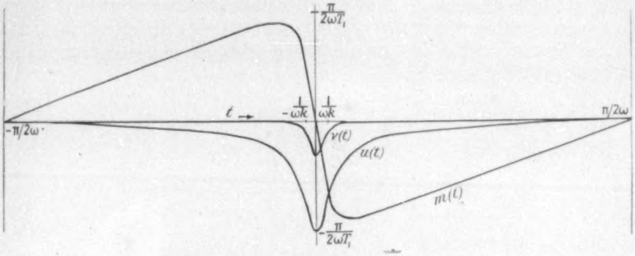

Figure 5. $2\pi \ll (\omega k)^{-1} \ll T_2$; $\omega T_1 \gg 1$.

We might expect that, if (17) is satisfied, the maximum deviation of $m(t)$ from its average value, the positive constant C, is small compared with C itself. We shall therefore replace $m(t+\tau)$ in (7) by this average value, i.e.

$$F(t) = -C\exp\{i(k/\omega)\cos\omega t\}\int_{-\infty}^{0} d\tau \exp\{\tau/T_2 - i(k/\omega)\cos\omega(t+\tau)\}. \qquad \ldots\ldots (28)$$

To obtain an approximation to $m(t)$ valid when (22) holds, we can express (8) in terms of integrals over a single sweep period and then expand the exponentials in powers of $2\pi/\omega T_1$. To second order in this expansion we get the following expressions:

$$m(n\pi/\omega)=1+(\omega T_1/\pi)\int_{-\pi/\omega}^{0} d\tau\, v(\tau)[1+\tau/T_1], \qquad \ldots\ldots(29)$$

$$m(t+n\pi/\omega)-m(n\pi/\omega)=\int_0^t d\tau\, v(\tau)-(\omega t/\pi)\int_0^{\pi/\omega} d\tau\, v(\tau), \qquad \ldots\ldots(30)$$

where n is any integer. Replacing $m(n\pi/\omega)$ in (29) by C we get a simple expression for C which, to first order in $2\pi/\omega T_1$, is

$$C=1+(\omega T_1/\pi)\int_0^{\pi/\omega} dt\, v(t). \qquad \ldots\ldots(31)$$

Since $v(t)$, as given by (28), is proportional to C, (31) is a linear equation determining C. For very large values of T_2 we shall actually find that the maximum value of (30) is *not* small compared with C. For this case however we shall find that the symmetry properties of $m(t)$ and $v(t)$ are such that equations (28) and (31) are still satisfied. Below we discuss the expressions obtained from (28)–(31) for various limiting values of T_2.

(i) $k\omega T_2^2 \ll 1$; $\omega T_2 \ll \pi$.

The expression (28) in this case reduces to C times the expressions given by (19) and (20), i.e.

$$u(t)=CT_2\{1+(kT_2\sin\omega t)^2\}^{-1}kT_2\sin\omega t, \qquad \ldots\ldots(32)$$

$$v(t)=-CT_2\{1+(kT_2\sin\omega t)^2\}^{-1}. \qquad \ldots\ldots(33)$$

Using (31) and (33) we find

$$C=\{1+T_1T_2[1+(kT_2)^2]^{1/2}\}^{-1}. \qquad \ldots\ldots(34)$$

Using (30) it can be shown that the maximum deviation of $m(t)$ from C is at most $C/k\omega$, which is indeed much less than C.

(ii) $1 \ll k\omega T_2^2$; $\omega T_2 \ll \pi$.

In this case T_2 is still small compared with the sweep period and $F(t)$ is given by C times the expression (21), i.e.

$$F(t)=\left.\begin{cases} -C\sqrt{\dfrac{2}{k\omega}}\exp\left[-\dfrac{ik\omega t^2}{2}\right]\left\{\Phi(\infty)-\Phi\left(\sqrt{\dfrac{k\omega t^2}{2}}\right)\right\} & \text{for } t<0, \\ -C\sqrt{\dfrac{2}{k\omega}}\exp\left[-\dfrac{ik\omega t^2}{2}-\dfrac{t}{T_2}\right]\left\{\Phi(\infty)+\Phi\left(\sqrt{\dfrac{k\omega t^2}{2}}\right)\right\} & \text{for } t>0. \end{cases}\right\} \qquad \ldots\ldots(35)$$

Equation (31) gives, after some calculation,

$$C=\{1+T_1/k\}^{-1}. \qquad \ldots\ldots(36)$$

The maximum deviation of $m(t)$ from C is again at most $C/k\omega$.

(iii) $T_2 \gg \pi/\omega$.

Using the periodicity condition (6) $F(t)$ as given by (28) can be re-written in terms of an integral over a single sweep period only. Using the fact that T_2 is much larger than the sweep period we obtain the following expression, to lowest order in an expansion in terms of $(\omega T_2)^{-1}$,

$$F(t)=-CT_2J_0(k/\omega)\exp\{i(k/\omega)\cos\omega t\}, \qquad \ldots\ldots(37)$$

where $J_0(x)$ is the Bessel Function of zero order which has the property that

$$\left.\begin{aligned} &J_0(x) \sim 1 && \text{for } x \ll 1, \\ &J_0(x) \sim (2/\pi x)^{1/2} \cos(x - \pi/4) && \text{for } x \gg 1. \end{aligned}\right\} \qquad \ldots\ldots (38)$$

The expression (37) shows that both $v(t)$ and $u(t)$ are symmetrical in the sense that $v(t) = v(-t)$ and $u(t) = u(-t)$. It then follows from (30) that $\{m(t) - m(0)\} = -\{m(-t) - m(0)\}$ and hence that $m(0)$ equals C, the average value of $m(t)$. This antisymmetry of $\{m(t) - C\}$ ensures that its contribution to the integral (7), which has been omitted in (28), vanishes to the approximation here considered even when $\{m(t) - C\}$ is not always smaller than C. The expression for C obtained from (37) and (31) is

$$C = \{1 + T_1 T_2 J_0{}^2(k/\omega)\}^{-1}. \qquad \ldots\ldots (39)$$

The maximum value of $\{m(t) - C\}$ is of the order of magnitude of $\{C T_2 J_0{}^2(k/\omega)/\omega\}$ which is not necessarily less than C if T_2 is sufficiently large. Sketches of v and u are given in Figures 6 and 7 for two different values of (k/ω).

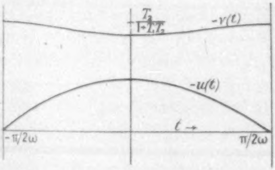

Figure 6. $2\pi\omega k \gg 1$; $\omega T_2 \gg 2\pi$; $k/\omega = \frac{1}{2}$.

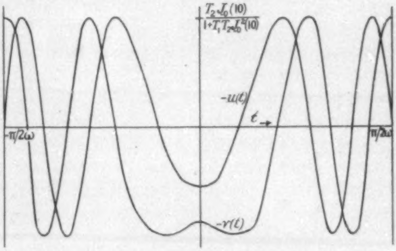

Figure 7. $2\pi\omega k \gg 1$; $\omega T_2 \gg 2\pi$; $k/\omega = 10$.

§ 5. DISCUSSION OF SIGNAL AMPLITUDES

In §§ 3 and 4 we have derived various approximate expressions for $m(t)$, $u(t)$ and $v(t)$, the three components of the resultant nuclear magnetic moment expressed as multiples of M_0, the thermal equilibrium value for the strong magnetic field H_0. Each of the formulae derived above is valid only when five parameters of the dimension of time (the two relaxation times, the resonance time, the sweep period and the Larmor period) satisfy certain extreme inequalities and the shape of the curves plotting these components against time is in general different for each of these cases. For some applications the quantity of main interest is the maximum value which a component of the nuclear moment in a direction perpendicular to that of the strong field H_z can assume. Expressed in units of M_0 this quantity is equal to the larger of the maxima of $u(t)$ and $v(t)$ and we shall denote it by $F_{\max}$. For each of the extreme cases treated in the above sections we can derive expressions for $F_{\max}$. For a number of these cases these expressions are similar enough to each other to be combined in a single empirical expression. Although such an expression has been proved to be a good approximation only for the limiting cases from which it was derived it seems likely that it is also at least a tolerable approximation in the intermediate regions.

Three such composite formulae are given below together with the range of validity claimed for them. We still assume the inequalities (2) and (9) to hold throughout. To facilitate comparison with experimental conditions we shall express all formulae in terms of τ_1, τ_2, τ_R, τ_L, τ_S (measured in seconds), γ, H_1 and H_S.

(i) $2\pi H_S/\gamma H_0 H_1 \tau_S = \tau_L/\tau_R \ll 1$; $(\tau_1/\tau_R), (\tau_2/\tau_R) \gg 1$.

$$F_{\max} = [1 + 4\tau_1/\tau_S]^{-1}. \qquad \ldots\ldots (40)$$

The case for which the relaxation times are smaller than the resonance time, for which $F_{\max}$ is found to be always less than unity, is not included. Apart from this, formula (40) embraces all cases of adiabatic passage whatever the values of τ_1 and τ_2. Note that for values of τ_1 larger than the sweep period, $F_{\max}$ is much smaller than unity however strong the inducing field H_1 is.

(ii) $2\pi H_S/\gamma H_0 H_1 \tau_S = \tau_L/\tau_R \gg 1$; $\tau_2/\tau_S \ll 1$.

$$F_{\max} = \left\{\frac{\tau_2 \gamma H_0}{1 + (2\tau_2^2 \gamma H_0 H_S/\tau_S H_1)^{1/2}}\right\}\left[1 + \frac{\tau_1 \tau_2 (\gamma H_0)^2}{(1 + [\tau_2 \gamma H_0 H_S/H_1]^2)^{1/2}}\right]^{-1}. \qquad \ldots\ldots (41)$$

Formula (41) embraces all cases of extremely non-adiabatic passage (sufficiently small H_1) for which τ_2 is much less than the sweep period. If, as is done in references A and B, the possibility of large values of τ_1 is neglected, then the term in square brackets in (41) (as well as the one in (40)) is replaced by unity. The largest value $F_{\max}$ can assume is $(\tau_S \gamma H_0 H_1/2H_S)^{1/2}$, which is much less than unity. Decreasing H_1 thus decreases the strength of the nuclear induction signals in these cases (but on the other hand increases their sharpness). If τ_1 is much larger than $(\tau_S \tau_L/\tau_R)$ or if τ_2 is much smaller than $(\tau_S \tau_L)^{1/2}$, then $F_{\max}$ is even smaller than $(\tau_S \gamma H_0 H_1/2H_S)^{1/2}$.

(iii) $2\pi H_S/\gamma H_0 H_1 \tau_S = \tau_L/\tau_R \gg 1$; $\tau_2/\tau_S \gg 1$.

$$F_{\max} = \tau_2 \gamma H_0 J_0(\tau_S \gamma H_0 H_S/2\pi H_1)[1 + \tau_1 \tau_2 (\gamma H_0)^2 J_0^2(\tau_S \gamma H_0 H_S/2\pi H_1)]^{-1}. \qquad \ldots\ldots (42)$$

Formula (42) covers the case where the sweep is so rapid that the sweep period is much less than both the relaxation times and the passage is extremely non-adiabatic. $F_{\max}$ increases again with increasing τ_2 and decreases with increasing τ_1. Since τ_1 can be much larger but no smaller than τ_2, the largest values are obtained for $F_{\max}$ if τ_1 is equal to τ_2. In this case $F_{\max}$ attains a value as large as $\frac{1}{2}$ whenever the following relation is satisfied:

$$\tau_2 \gamma H_0 J_0(\tau_S \gamma H_0 H_S/2\pi H_1) = 1. \qquad \ldots\ldots (43)$$

This result may seem somewhat surprising since it implies that nuclear induction signals of equal strength can be induced by an arbitrarily weak rotating field H_1 and by a reasonably strong one. But (43) shows that, for a very weak field H_1, signals of such large strength can be obtained only if τ_1 and τ_2 are equal and very large $(\tau_2/\tau_S \gtrsim (\tau_L/\tau_R)^{1/2} \sim (H_S/\gamma H_0 H_1 \tau_S)^{1/2})$ and it then takes very many sweep periods after H_1 is switched on for the signals to build up to this strength.

Another interesting consequence of (42) is the critical dependence of the signal strength on the exact value of $(\tau_S \gamma H_0 H_S/H_1)$ for large values of this quantity, since $J_0(x)$ is an oscillating function for large x. The expression (37), and hence (42), represents only the first term in an expansion in powers of (τ_S/τ_2) and hence the amplitude of the induced signals will not vanish exactly even when $J_0(\tau_S\gamma H_0 H_S/2\pi H_1)$ is zero. Nevertheless the signal amplitudes will vary considerably if the value of $(\tau_S\gamma H_0 H_S/2\pi H_1)$ is changed by $\pi/2$, however small a fractional change of τ_S or H_S this change may represent.

GLOSSARY OF SYMBOLS USED

γ = gyromagnetic ratio
p = angular frequency of rotating (radio-frequency) field
H_1 = amplitude of rotating field
$H_z(t)$ = field in the z-direction
H_S = sweep amplitude of $H_z(t)$
H_0 = resonance value of $H_z(t) = p/\gamma$
τ_L = period of Larmor precession $= 2\pi/\gamma H_0$
τ_S = repetition period of the sweep of $H_z(t)$
τ_R = resonance time $= \tau_S H_1/H_S$
τ_1 = longitudinal relaxation time
τ_2 = transverse relaxation time
t = (time in seconds) × (γH_0)
M_0 = thermal equilibrium nuclear moment
$m(t)$ = (nuclear moment in z-direction)$/M_0$
$u(t)$ = (in-phase component of moment)$/M_0$
$v(t)$ = (out-of-phase component of moment)$/M_0$
$F(t)$ $= v(t) + iu(t)$
$F_{\max}$ = larger of the maxima of $u(t)$ and $v(t)$

$T_S = \tau_S\gamma H_0$, $T_R = \tau_R\gamma H_0$
$T_1 = \tau_1\gamma H_0$, $T_2 = \tau_2\gamma H_0$
$\omega = 2\pi/T_S$, $k = H_S/H_1$

ACKNOWLEDGMENTS

The author wishes to thank Dr. J. S. Gooden for helpful discussions and the Department for Scientific and Industrial Research for a Senior Research Award.

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γ-Decay and Mesons of Spin Zero

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Communicated by R. E. Peierls; MS. received 30th August 1949

ABSTRACT. On the assumption that τ-mesons are scalar particles and have an appreciable interaction with nucleons, the lifetime of a τ-meson at rest decaying into two photons and a pseudoscalar π-meson, is calculated by means of standard perturbation theory. The lifetime obtained is $G \times 10^{-12}$ sec. where G is the reciprocal of the product of the dimensionless meson–nucleon coupling parameters. This result is roughly what can be expected from observations and, taken together with the results of a previous paper, seems to indicate that the heavy mesons have spin zero.

§ 1. INTRODUCTION

It has been reported (for a recent list of references see Powell *et al.* 1949) that mesons with a mass of about 900 electron masses * (usually called τ-mesons) are produced by cosmic rays. On the other hand π-mesons with mass about 300 electron masses have been produced in the Berkeley experiments as well as by cosmic rays. If these production processes are regarded as the consequence of the elementary process

$$\text{proton} \rightarrow \text{neutron} + \text{positive meson}$$

and similar processes, the immediate conclusion to be drawn from these observations is that in both cases the nucleon–meson coupling parameter is not very small.

Consequently τ-mesons must be fairly unstable towards decay into π-mesons and photons, where the decay process takes place by means of virtual creation and annihilation of nucleons.

The standard perturbation calculations for the most probable case where only one photon is emitted have been carried out by the present author (van Wyk 1949, in the following referred to as I). These calculations cover all possibilities for mesons of spin 0 and 1 except those for which both the initial and final mesons have spin zero. The emission of one photon in the excepted cases is forbidden by the conservation of angular momentum.

In the present paper the lifetime of a scalar meson at rest, decaying into a pseudoscalar meson and two photons, is calculated by means of perturbation theory. While the lifetimes derived in I are very short, the result of the present calculation is fairly satisfactory and the lifetime is $G \times 10^{-12}$ sec. where G is the reciprocal of the product of the dimensionless meson–nucleon coupling parameters.

The lifetime for the case with scalar and pseudoscalar mesons interchanged is about the same while the cases where both mesons have the same tensor character lead to divergent integrals.

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* According to private communications from Professor Powell and Dr. Peters the evidence for the existence of τ-mesons in large numbers is now in doubt.

§ 2. MATRIX ELEMENT

Consider the decay of a positive scalar meson, mass μ_0, at rest into a positive pseudoscalar meson, mass μ_3 momentum p_3, and two γ-rays momenta $p_i (i=1, 2)$ according to the scheme

$$\mathrm{M}_s^+(0) \rightarrow \mathrm{m}_{ps}^+(p_3) + \gamma(p_1) + \gamma(p_2).$$

Using units in which $\hbar = c = 1$, the conservation of energy and momentum requires

$$\mu_0 = \epsilon_3 + p_1 + p_2, \qquad \ldots\ldots(1\,a)$$

$$0 = \mathbf{p}_1 + \mathbf{p}_2 + \mathbf{p}_3, \qquad \ldots\ldots(1\,b)$$

where $\epsilon_3 = (\mu_3{}^2 + p_3{}^2)^{\frac{1}{2}}$.

The decay process is assumed to take place by means of virtual creation and annihilation of nucleons. A typical way in which this process can take place is the following:

Process	Matrix element
$M^+(0) \rightarrow P^+(q) + N^-(-q)$	$a^*(q) X b(-q)$
$P^+(q) \rightarrow P^+(q_1) + \gamma(p_1)$	$a^*(q_1) Y a(q)$
$P^+(q_1) \rightarrow P^+(q_{12}) + \gamma(p_2)$	$a^*(q_{12}) Z a(q_1)$
$N^-(-q) + P^+(q_{12}) \rightarrow m^+(p_3)$	$b^*(-q) T a(q_{12})$

where the notation used is the same as in I, and

$$\mathbf{q}_i = \mathbf{q} - \mathbf{p}_i, \qquad \mathbf{q}_{ij} = \mathbf{q}_i - \mathbf{p}_j. \qquad \ldots\ldots(2)$$

According to perturbation theory, the compound matrix element for the four stage process written out above, is

$$\int n_{0123}[(-2\epsilon_q)^3 d_{0123}]^{-1} d^3q,$$

where d_{0123} will be discussed later, and

$$n_{0123} = \tfrac{1}{16} \mathrm{Sp}\, T(1+E_{12}) Z(1+E_1) Y(1+E) X(1-E).$$

For the sake of simplicity only the g terms in I, viz. the interaction terms independent of the derivatives of the field variables, will be used. Therefore

$$X = \beta, \qquad Y = (\alpha e_1),$$

$$Z = (\alpha e_2), \qquad T = \beta\gamma_5, \qquad (\gamma_5 = -i\alpha_1\alpha_2\alpha_3).$$

Putting $W = \gamma_5(\alpha v)$, it follows that $n_{0123} = \frac{1}{8} \mathrm{Sp}\, W(1+E_{12}) Z(1+E_1) Y(1+E)$.

The four stages of the decay process can be permuted in 24 ways. In four of these permutations the two γ-rays are emitted in succession—by a proton, say—in the first two stages. The theory is symmetrical between protons and antiprotons as regards emission of photons; hence four new permutations are added if an antiproton emits the two γ-rays. Similarly four more new ones are added when the two γ-rays are emitted in the last two stages. A neutron—if its magnetic moment is neglected—cannot emit a photon and a proton cannot absorb a positive meson. Hence there exist no further symmetries which can give rise to new ways. The total number of ways in which the decay process can take place is therefore 32($=24+4+4$).

The matrix element that leads directly to the lifetime of the initial meson is

$$H=(2\pi/L^3)^2e^2g_0g_3(\mu_0p_1p_2\epsilon_3)^{-\frac{1}{2}}(L/2\pi)^3d^3\int q\sum_{32}n[(-2\epsilon_q)^3d]^{-1}, \quad \ldots\ldots(3)$$

where L is the length of a side of the cubic box in which the system is supposed to be enclosed and the summation is over the total number of processes.

Because of the similarity between (αe_1) and (αe_2) the total number of ways can be divided into two equal groups of 16 processes each in such a way that the contribution to H of the second group is derived from that of the first group by permuting e_1 and e_2. It is therefore only necessary to calculate the contribution of a set S of 16 suitably chosen ways.

From a detailed calculation it follows that

$$\begin{aligned}
n_{0123}&=\tfrac{1}{8}\mathrm{Sp}\,W(1+E_{12})Z(1+E_1)Y(1+E) &&=-n_{3210},\\
n_{0132}&=-\tfrac{1}{8}\mathrm{Sp}\,W(1-E_{12})Z(1+E_1)Y(1+E) &&=-n_{2310},\\
n_{0312}&=-\tfrac{1}{8}\mathrm{Sp}\,W(1-E_{12})Z(1+E_1)Y(1+E) &&=-n'_{2130},\\
n'_{0321}&=\tfrac{1}{8}\mathrm{Sp}\,W(1-E_{12})Z(1-E_1)Y(1+E) &&=-n_{1230},\\
n_{1032}&=-\tfrac{1}{8}z\,\mathrm{Sp}\,T(1-E_{12})Z(1+E_1)Y(1-E) &&=-n_{2301},\\
n_{1023}&=\tfrac{1}{8}z\,\mathrm{Sp}\,T(1+E_{12})Z(1+E_1)Y(1-E) &&=-n_{3201},\\
n_{1203}&=\tfrac{1}{8}z\,\mathrm{Sp}\,T(1+E_{12})Z(1+E_1)Y(1-E) &&=-n'_{3021},\\
n_{3012}&=\tfrac{1}{8}z\,\mathrm{Sp}\,T(1-E_{12})Z(1+E_1)Y(1+E) &&=-n'_{2103},
\end{aligned}$$

where an accent denotes the emission of the photons by the antiproton. The 16 expressions written down form a set S.

In the cases where relation

$$-n_{ijkl}=n_{lkji} \quad \ldots\ldots(3\,a)$$

is not evident it can easily be proved as in the following example:

$$\begin{aligned}
n_{3210}&=-\tfrac{1}{8}\mathrm{Sp}\,W(1-E_{12})Z(1-E_1)Y(1-E)\\
&=-\tfrac{1}{8}\mathrm{Sp}\,T\{W(1-E_{12})Z(1-E_1)Y(1-E)\}T^{-1}\\
&=-n_{0123}
\end{aligned}$$

since T anticommutes with α and β.

From the list above it follows that

$$\sum_{16}n_{ijkl}=0, \quad \ldots\ldots(4)$$

where $\sum_{16}$ denotes summation over S.

Expand in powers of $x=\mu_0/M$. Then

$$n={}^0n+x\,{}^1n+x^2\,{}^2n+\ldots,$$

$$d^{-1}=1+x\,{}^1d+x^2\,{}^2d+\ldots.$$

The term ${}^0n$ is obtained by replacing E_i by E and

$${}^0n_{ijkl}=0. \quad \ldots\ldots(5)$$

Hence by (4) and (5)

$$\sum_{16} n/d = \sum_{16} \{{}^0n + x({}^0n\,{}^1d + {}^1n) + x^2({}^0n\,{}^2d + {}^1n\,{}^1d + {}^2n) + \ldots\}$$

becomes

$$\sum_{16} n/d = x^2 \sum_{16} {}^1n\,{}^1d. \qquad \ldots\ldots(6)$$

The expressions ${}^1n$ are calculated by using the expansions

$$E_1 = E + xz\{(vl_1)E - (\alpha l_1)\} + \ldots,$$
$$E_{12} = E - xz\{(vl_3)E - (\alpha l_3)\} + \ldots,$$

where $l_i = p_i/\mu_0$.

$$8\,{}^1n_{0123} = z\,\mathrm{Sp}\, W(1+E)Z\{(vl_1)E - (\alpha l_1)\}Y(1+E)$$
$$- z\,\mathrm{Sp}\, W\{(vl_3)E - (\alpha l_3)\}Z(1+E)Y(1+E).$$

Anticipating the fact that ${}^1d$ are effectively only dependent on $|v|$, the simplification of integrating over the directions of **v** can be introduced at this stage. Hence

$$(1/4\pi)\int {}^1n_{0123}\,dw = -(2i/3)zv^2(e_{1\wedge}e_2)l_1 - (i/3)zv^2(e_{1\wedge}e_2)l_3.$$

Similarly

$$(1/4\pi)\int {}^1n_{0132}\,dw = -(i/3)zv^2(e_{1\wedge}e_2)l_3$$
$$= (1/4\pi)\int {}^1n_{0312}\,dw = (1/4\pi)\int {}^1n_{1230}\,dw,$$
$$(1/4\pi)\int {}^1n_{1032}\,dw = iz^3(e_{1\wedge}e_2)l_3,$$
$$(1/4\pi)\int {}^1n_{1023}\,dw = iz^3(e_{1\wedge}e_2)l_1 + iz^3(e_{1\wedge}e_2)l_3$$
$$= (1/4\pi)\int {}^1n_{1203}\,dw,$$
$$(1/4\pi)\int {}^1n_{3012}\,dw = -iz^3(e_{1\wedge}e_2)l_1.$$

Before evaluating ${}^1d$ it is useful to notice that the part of $\sum_{16} {}^1n\,{}^1d$ odd in **v** vanishes on integration over the angles of v. As shown above, ${}^1n$ are even in **v**, hence it is only necessary to determine that part of ${}^1d$ even in **v**.

A general expression for the relevant part of ${}^1d_{ijkl}$ can be obtained easily by considering a special case. The expansion

$$\epsilon_{q_1} = \epsilon_q\{1 - xz(vl_1) + \ldots.\} \qquad \ldots\ldots(6\,a)$$

has been derived in I. Since only terms of order x and even in **v** need be considered for the calculation of ${}^1d$, ϵ_{q_1} can henceforth be replaced by ϵ_q.

Therefore

$$(-2\epsilon_q)^3\,d_{0123} = (\mu_0 - 2\epsilon_q)(\mu_0 - \epsilon_{q_1} - \epsilon_q - \epsilon_1)(\mu_0 - \epsilon_q - \epsilon_{q_{12}} - \epsilon_1 - \epsilon_2)$$
$$\simeq (\mu_0 - 2\epsilon_q)(\mu_0 - 2\epsilon_q - \epsilon_1)(\mu_0 - 2\epsilon_q - \epsilon_1 - \epsilon_2)$$
$$\simeq (-2\epsilon_q)^3(1 - \tfrac{1}{2}xz\theta_0)\{1 - \tfrac{1}{2}xz(\theta_0 - \theta_1)\}\{1 - \tfrac{1}{2}xz(\theta_0 - \theta_1 - \theta_2)\}$$

or

$$d_{0123}^{-1} \simeq 1 - \tfrac{1}{2}xz(-3\theta_0 + 2\theta_1 + \theta_2) + \ldots.$$

This can be generalized to

$$-{}^{1}d_{ijkl}=(z/2)(3\theta^{i}+2\theta^{j}+\theta^{k}), \qquad \ldots\ldots(7)$$

where $\theta^{0}=-\theta_{0}$, $\theta^{i}=\theta_{i}\,(i=1,2,3)$.
The right hand side of (6) can now be calculated and the result integrated over the magnitudes of the momenta of the intermediate nucleons.

$$\begin{aligned}-\int d^{3}q\sum_{16}n[(-2\epsilon_{q})^{3}d]^{-1}\\ &=(8\pi i/8\,.\,15)x^{2}(e_{1\wedge}e_{2})l_{1}(2+3\theta_{1}+2\theta_{2}-3\theta_{3})\\ &\quad+(4\pi i/8\,.\,15)x^{2}(e_{1\wedge}e_{2})l_{3}(2+7\theta_{1}-5\theta_{3}). \qquad \ldots\ldots(8)\end{aligned}$$

The contribution of the second group of 16 processes is obtained by interchanging the suffixes 1 and 2 in (8).

Using the relation (1 *b*) expressing the conservation of momentum, the total contribution from all 32 processes becomes

$$(4\pi x^{2}/8\,.\,15)Q$$

with $$Q=2i(e_{1\wedge}e_{2})l_{1}(9-11\theta_{3})+i(e_{1\wedge}e_{2})l_{3}(3+12\theta_{1}-5\theta_{3}).$$

For given p_1 and p_3 the matrix element (3) is therefore

$$H=(2\pi/L^{3})^{2}\,e^{2}g_{0}g_{3}(\mu_{0}p_{1}p_{2}\epsilon_{3})^{-\frac{1}{2}}(4\pi x^{2}/8\,.\,15)Q(L/2\pi)^{3}. \qquad \ldots\ldots(9)$$

The justification for taking only the leading term into account follows from (3 *a*). The proof is similar to the one given in I.

§ 3. ADDITIONAL PROCESSES

The processes 7 .... 12 in I describe ways of decay in which the photon is emitted by the meson and not by the nucleons. These processes will also contribute in the present problem if the meson emits the two photons simultaneously instead of the single photon as in I. This transition is made possible by the term in the meson–electromagnetic interaction energy which is quadratic in both the meson and photon variables. The contribution from these processes to the total matrix element will then contain as a factor the matrix element for the process

$$\mathrm{M}_{s}^{+}\rightarrow\mathrm{P}^{+}+\mathrm{N}^{-}\rightarrow\mathrm{m}_{ps}^{+}$$

which is forbidden by the conservation of parity. Hence these processes do not contribute to the decay of a scalar τ-meson into a pseudoscalar π-meson and two photons.

The processes 13 .... 16 contribute only to *f*-terms, and therefore the total contribution is given by (9).

§ 4. THE LIFETIME

The lifetime τ of the initial meson is given by

$$\tau^{-1}=2\pi\int\Sigma|H|^{2}\rho\,dp_{1}\,dp_{3}\,d\omega_{3}, \qquad \ldots\ldots(10)$$

where $\rho\,dp_1\,dp_3\,d\omega_3$ is the number of final states per unit energy and the summation is over the polarizations of the two photons.

With p_2 and μ_3 given, the equations (1) specify an ellipse with foci at the extremities of p_3 and with major axis

$$p_{1}+p_{2}=2\eta.$$

If further $|p_1|(|p_2|)$ is given, $|p_2|(|p_1|)$ is determined by the conservation laws. Hence the number of final states is given by

$$\rho_0\, dE = f\rho_{p_3}\, d\epsilon_3 \rho_{p_1}\, dp_1, \qquad \ldots\ldots(11)$$

where

$$\rho_{p_3}\, d\epsilon_3 = p_3^2\, dp_3\, d\omega_3 (L/2\pi)^3, \qquad \ldots\ldots(12)$$

and f is a statistical factor.

Suppose (x, y) are Cartesian coordinates in the plane of the ellipse so that it is given by

$$x^2/\eta^2 + y^2/(\eta^2 - b^2) = 1, \qquad \ldots\ldots(13)$$

where $p_3 = 2b$. The circle centre $(b, 0)$ and radius p_1 is given by

$$(x - b)^2 + y^2 = p_1^2. \qquad \ldots\ldots(14)$$

Alternatively (η, p_1) instead of (x, y) can be regarded as coordinates in the plane of the ellipse. The number of states for which p_1 lies between p_1 and $p_1 + dp_1$ is determined by the intersection of the shells $(p_1, p_1 + dp_1)$ and $(\eta, \eta + d\eta)$. This is given by the volume of the body of revolution with section

$$J\left(\frac{x, y}{\eta, p_1}\right) d\eta\, dp_1,$$

its volume being

$$2\pi y J\left(\frac{x, y}{\eta, p_1}\right) d\eta\, dp_1.$$

By means of (13) and (14) it is easy to show that

$$2\pi y J\left(\frac{x, y}{\eta, p_1}\right) d\eta\, dp_1 = 2\pi \frac{p_1 p_2}{p_3} d(2\eta)\, dp_1$$

so that

$$\rho_{p_1}\, dp_1 = 2\pi \frac{p_1 p_2}{p_3} d(p_1 + p_2)\, dp_1 \left(\frac{L}{2\pi}\right)^3. \qquad \ldots\ldots(15)$$

Hence from (11), (12) and (15)

$$\begin{aligned}\rho_0 &= \rho\, dp_1\, dp_3\, d\omega_3 \\ &= \pi (L/2\pi)^6 p_1 p_2 p_3\, dp_1\, dp_3\, d\omega_3,\end{aligned} \qquad \ldots\ldots(16)$$

where the factor $f = \frac{1}{2}$ comes from the indistinguishability of the two photons.

By (9) and (16), (10) becomes

$$\tau^{-1} = 2/[8^2(15)^2(2\pi)^4] e^4 g_0^2 g_3^2 x^4 \int (p_3/\mu_0\epsilon_3) \Sigma |Q|^2\, dp_1\, dp_2\, d\omega_3,$$

with the following intervals of integration:

$$\tfrac{1}{2}\mu \geqslant p_1 \geqslant \tfrac{1}{2}\mu - p_3,$$

$$(\mu_0^2 - \mu_3^2)/2\mu_0 \geqslant p_3 \geqslant 0,$$

where

$$\mu = \mu_0 - \epsilon_3 + p_3.$$

Introduce $l_1 = p_1/\mu_0$, $y = (\mu_3^2 + p_3^2)^{\frac{1}{2}}/\mu_0$, $a = \mu_3/\mu_0$, then

$$\tau^{-1}(16(15)^2(2\pi)^3)^{-1} e^4 g_0^2 g_3^2 \mu_0 x^4 \int \Sigma |Q|^2\, dl_1\, dy \qquad \ldots\ldots(17)$$

with the intervals

$$\tfrac{1}{2}(1 - y + s) \geqslant l_1 \geqslant \tfrac{1}{2}(1 - y - s)$$

$$\tfrac{1}{2}(1 + a^2) \geqslant y \geqslant a,$$

where $s = l_3 = p_3/\mu_0 = (y^2 - a^2)^{\frac{1}{2}}$. Summation over the photon polarization gives

$$\begin{aligned}\Sigma|Q|^2 = {} & 4(1+\cos^2\omega_{12})l_1^2(9-11y)^2 \\ & +(\cos^2\omega_{23}+\cos^2\omega_{13})l_3^2(3+12l_1-5y)^2 \\ & +4(-\cos\omega_{13}+\cos\omega_{23}\cos\omega_{12})l_1l_3(9-11y)(3+12l_1-5y),\end{aligned}$$

where ω_{ij} is the supplement of the angle between l_i and l_j. Expressing the cosines in terms of s, $(1-y)$ and l_1 by means of the usual relations holding for a triangle, the integration over l_1 and y can be carried out so that finally

$$\begin{aligned}f(a) &= \tfrac{1}{2}\int dy \int dl_1 \Sigma|Q|^2 \qquad \ldots\ldots(18) \\ &= 1{\cdot}5641 + (49{\cdot}0302 + 40{\cdot}5\log a)a^2 \\ &\qquad + (0{\cdot}8505 + 128{\cdot}75\log a)a^4 \\ &\qquad - (51{\cdot}4916 - 38{\cdot}3956\log a)a^6 + \ldots.\end{aligned}$$

Hence, by (17) and (18),

$$\tau^{-1} = [8(15)^2(2\pi)^3]^{-1}e^4 g_0^{\,2} g_3^{\,2} \mu_0 x^1 f(a).$$

With $x = \frac{1}{2}$ and taking the mass of the π-mesons as 286 electron masses, $a = \mu_3/\mu_0 = 0{\cdot}3112$ and $f(0{\cdot}3112) = 0{\cdot}243$; hence in the usual units

$$\tau^{-1} = 1{\cdot}338 \times 10^{12} g_0^{\,2} g_3^{\,2}\,\text{sec}^{-1}. \qquad \ldots\ldots(19)$$

The result (19) refers only to the nucleon–meson interaction terms independent of the derivatives of the meson field variables. These terms are very simple and the relations (4) and (5) which lead to the small value on the right hand side of (6) are due to this simplicity.

The three remaining coupling combinations, with one or both couplings referring to derivative-dependent interactions, are more complicated and will not lead to expressions smaller than the right-hand side of (6). In any case, provided they are finite, the expressions derived from these coupling combinations corresponding to (8) will differ effectively from (8) only by a factor x^{-1} or x^{-2}, which for $x = \frac{1}{2}$ is not very large.

Hence (19) can be taken to be representative of all four lifetimes.

§ 5. ALTERNATIVE MESON COMBINATIONS

The decay scheme

$$\mathrm{M}_{\mathrm{ps}}^{+}(0) \rightarrow \mathrm{m}_{\mathrm{s}}^{+}(p_3) + \gamma(p_1) + \gamma(p_2)$$

also leads to the relations (4), (5) and (6) and therefore the result (19) also holds good for this case.

However if both mesons have the same tensor character, the expressions corresponding to (8) are logarithmically divergent, and the processes discussed in § 3 are no longer forbidden by the conservation of parity but lead to quadratic divergences.

§ 6. PAIR CREATION

The matrix element (9) is of the order α(= fine structure constant) and relates to γ-decay. Another effect in connection with meson decay which is also of the order α is that of the creation of an electron–positron pair. The 'mechanism' of this effect can be thought of as follows: By their Coulomb interaction an electron and positron are created, the energy being supplied by the process

$$M^+(0) \rightarrow P^+(q) + N^-(-q),$$
$$P^+(q) + N^-(-q) \rightarrow m^+(0).$$

Hence the compound matrix element of the pair creation decay process contains as a factor expressions discussed in § 3. Hence transitions which are accompanied by a change in parity are forbidden, while, as mentioned in § 5, quadratic divergences occur for transitions without change in parity.

§ 7. CONCLUSION

The results of the present investigation combined with those of I seem to support the view that both τ- and π-mesons have spin zero and opposite parity. However these results are derived on the assumptions that (*a*) the reaction $P^+ \rightarrow N + \tau^+$ is the fundamental one for a system of nucleons and τ-mesons; (*b*) the results derived from ordinary perturbation theory will not be greatly affected by a future more satisfactory theory.

The validity of the conclusion that both τ- and π-mesons have spin zero depends therefore on the validity of the assumptions (*a*) and (*b*), as well as on the evidence for the frequent occurrence of τ-mesons.

ACKNOWLEDGMENTS

The stimulating interest and criticism of Professor R. E. Peierls and grants from the South African Council for Scientific and Industrial Research and the University of Birmingham, are gratefully acknowledged.

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On the Velocity of Discharge Propagation in Externally Quenched Geiger Counters

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MS. received 2nd August 1949

ABSTRACT. A new method for measuring the propagation velocity of the discharge along a Geiger non-proportional counter is described. The results obtained with cylinder grade argon, hydrogen and neon are summarized graphically, and a brief discussion of propagation mechanisms is given.

§ 1. INTRODUCTION

THE space charge sheath formed round the wire of a conventional wire-cylinder Geiger tube counter initially at a point determined by the first radial avalanche due to the counted particle, spreads along the counter towards each end, provided that the operating voltage of the tube is sufficiently high. The mechanism by which the discharge spreads has been studied by many authors. It appears reasonable to assume that, since photo-emission from the cathode is relatively negligible in the case of self-quenched counters (Stever 1942), the discharge in such a counter spreads along the wire (as indicated by the experiments of Stever (1942), Wilkening and Kanne (1942) and others on beaded wire counters). This occurs presumably by a series of photo-ionization processes, each followed by a short radial Townsend avalanche (see (*a*), Figure 1). The mechanism of production of these photo-ionizing radiations has been discussed by Hopwood (1949). The velocity of propagation of the discharge for this case has been determined by van Gemert *et al.* (1942) and, more directly, by Hill and Dunworth (1946), Huber *et al.* (1946), Alder *et al.* (1947), Wantuch (1947), Freeman (see Wilkinson 1948), and finally by Knowles *et al.* (1948). In the case of externally quenched counters i.e. those containing the elementary gases such as hydrogen, argon, etc., the only published measurements appear to be those of

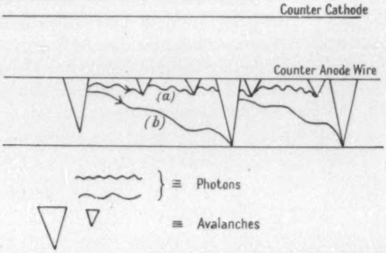

Figure 1. Mechanisms of discharge spread in a Geiger counter.

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‡ Now returned to India.

Knowles *et al.* (1948), to which those of the present paper constitute a sequel. Since absorption of the energetic photons produced in the primary avalanche is not now so likely, as shown by the experiments of Craggs and Jaffe (1947 a, b), an appreciable photo-emission from the cathode cylinder is probable, and the mechanism (*b*) of Figure 1 may be operative. Corson and Wilson (1948) in their excellent review article on counters, also point this out, and suggest that the resulting velocity of discharge propagation should be very high, although they could not quote data.

Measurements of the velocity of discharge propagation are of interest for several reasons. They explain and define certain limitations to coincidence counting, as first pointed out by Dunworth (1939). Also they might, if mechanism (*a*) in Figure 1 is operative, provide useful data on photo-ionization processes (Jaffe *et al.* 1949) needed for investigations of streamer discharges (Meek 1940, Loeb and Meek 1941, Raether 1940, Hopwood 1949), and they lead to the development of new counter techniques (Frisch 1949). The effect of the finite velocity of propagation on Geiger pulse shapes has been described by van Gemert *et al.* (1942), Corson and Wilson (1948) and others.

The purpose of the present paper, therefore, is to report detailed measurements with hydrogen, argon and neon, and to discuss the mechanisms illustrated in Figure 1.

§ 2. THE EXPERIMENTAL METHOD

The technique developed in these experiments differs from those described by earlier authors and the circuit used is shown schematically in Figure 2. A pulse resulting from discharge build-up in one of the short end cylinders (the 'start' cylinder) of the long counter tube shown (1 or 2 m. in length) connects a 4 Mc/s. quartz-controlled oscillator through a gate circuit to a fast scaler and recorder. The discharge which spreads along the counter in both directions from the point of initiation, and which we assume for the present to start from a point very near

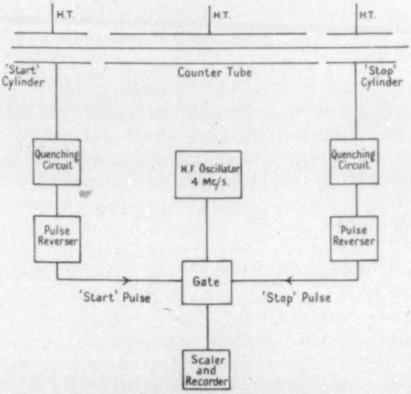

Figure 2. Schematic circuit diagram.

to the start cylinder, will later reach the 'stop' cylinder, and the pulse produced therein closes the gate. The intervening number of oscillations, each 0·25 μsec. in duration, is indicated by the recorder.

The circuits were designed and adjusted by A. J. Knowles, and will be described in detail by him in a separate publication. The following is a necessary but brief account included here for the sake of completeness.

The quenching circuits were of the conventional Neher–Harper type, and the pulse reversing circuits possess no features of special interest. The crystal-controlled oscillator is of the cathode follower type (Schlesinger 1945), possesses a low output impedance, and provides an output of about 25 v. peak of saw-tooth form which is especially suitable for use with scaling circuits. The gate circuit (Knowles 1950) operates with about 0·2 μsec. delay, constant for either start or stop operation. A scaling factor of 400 : 1 was used, with two scale-of-two stages followed by two scale-of-ten stages, and adequate resolution was obtained by using valves with very low inter-electrode capacities and germanium crystal diodes to remove overshoot on the grid coupling condensers at the switch-over. These circuits were used up to a scaling frequency of 8 Mc/s.

The counter tubes were of conventional design with tungsten wires 5 mil in diameter, and copper cathodes 0·75 inch diameter. One and two metre counters with single cylinders and short (10 cm.) end cylinders were usually used, but some results were also obtained with a 2 m. counter in which ten 20 cm. cylinders were mounted, so that the operative counter length could be changed easily without alteration of the gas-filling. Great care was taken in the alignment of the cylinders, each of which was supported by an adequate number of stout tungsten leads sealed through the C9 glass tubes. It was not possible to bake such long tubes, but they were carefully cleaned with chromic acid before use (for this purpose it is convenient to provide a wide bore filling tube at each end of the counter). The following gases of cylinder purity were used: hydrogen (about 0·5% oxygen), argon (about 1% nitrogen) and neon (about 2% helium).

§ 3. EXPERIMENTAL RESULTS

The circuits were first checked with a pulse generator, and with a special commutator device which enabled sharp pulses, with variable interval, to be produced. The scaling circuits, as already stated, were proved to count satisfactorily at frequencies up to 8 Mc/s. In order to check the whole circuit of Figure 2 still further, some measurement of the propagation velocity in an alcohol–argon filled counter were made for comparison with the results of Hill and Dunworth (1946) Alder *et al.* (1947), (extrapolated), and Miss Freeman (see Wilkinson 1948). The results of Wantuch (1947) could not be directly compared, since the required values of overvoltage are lacking; the available data are plotted in Figure 3 to which the details given in Table 1 are applicable. In these experiments the quenching circuits (Figure 2) were not used.

The data of Alder *et al.*, used in Figure 3, has been obtained by extrapolation from various alcohol partial pressures, down to 10 mm., to the required value of 5 mm. It is unfortunate that their measurements were all taken with a total pressure of 8 cm. Hg, so that direct comparison with the other results of Figure 3 is impossible. It is not surprising that the results of Alder *et al.* (with a relatively higher alcohol content) fall below those of other workers (see also Hill and Dunworth 1946). Other slight differences in the experimental conditions are enumerated in Table 1 but their effects are likely to be so slight as not seriously to affect the good agreement shown in Figure 3 where the difference in velocities is only apparent at high overvoltages, where it is still possible that the differences shown in Table 1 may be more important.

The type of transit time spectrum obtained with the argon–alcohol counters was of the general form shown, for example, in Figure 7.

The results obtained with 'slow' counter fillings, i.e. elementary gases without the admixture of a polyatomic quenching agent, are shown in Figures 4 to 6. The background counts were so high with these fillings that most of the discharges started in the long middle cylinder, and of course travelled in two opposing directions along the tube. The resultant transit times, measured between the times of appearance of pulses in the start and stop cylinders of Figure 2, show a spectrum from zero time to a maximum which is the time taken for a discharge to travel the full length of the counter. The start cylinder was operated at a higher overvoltage than the middle cylinder in order to increase to a maximum the number of discharges initiated in the start cylinder. (The term overvoltage denotes the difference between the operating voltage and the starting voltage; the latter is defined below.)

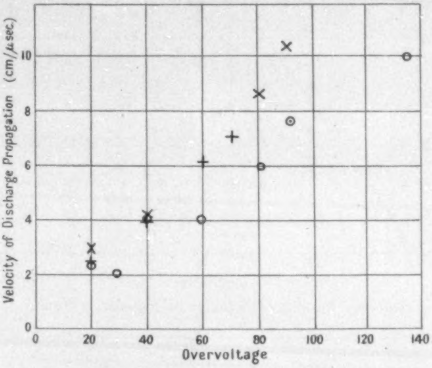

Figure 3. Experimental data on propagation velocities.
(9·5 cm. Hg argon, 0·5 cm. Hg ethyl alcohol)

○ Alder *et al.*
× Hill and Dunworth.
+ Freeman.
⊙ Knowles *et al.* (points at 20, 40 and 92 v.).

Table 1

Authors	Counter cathode diam.	Counter wire diam.	Gas filling, partial pressures (cm. Hg)	
			argon	ethyl alcohol
Alder *et al.*	1·8 cm.	0·15 mm.	7·5	0·5
Hill and Dunworth	0·75 in.	0·008 in.	9·5	0·5
Freeman	—	—	9·5	0·5
Present authors	0·75 in.	0·005 in.	9·5	0·5

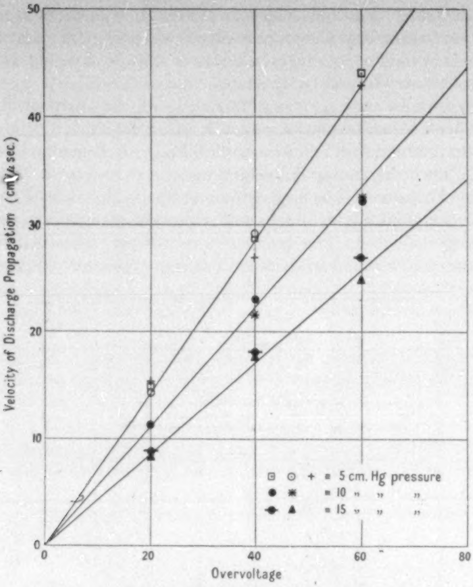

Figure 4. Velocity of discharge propagation in argon.

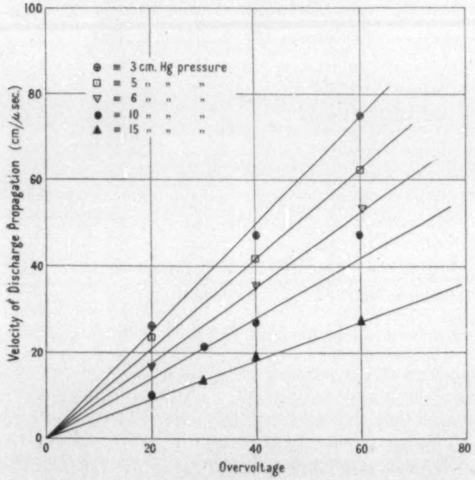

Figure 5. Velocity of discharge propagation in hydrogen.

A typical transit time spectrum (taken with argon) is shown in Figure 7, where the numbers of observed transits for successive intervals of 0·25 μsec. are plotted. Each point in the curves of Figures 4 to 6, which show the tabulated results for argon, hydrogen and neon respectively, represents at least one such spectrum, and each spectrum (see Figure 7) includes many hundreds of readings. The results were checked over a period of almost a year and showed consistency. Further, the velocities of propagation measured with different effective counter lengths in the same gas filling agreed within the errors of measurement, thus

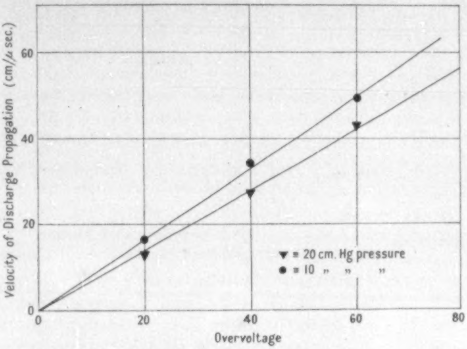

Figure 6. Velocity of discharge propagation in neon.

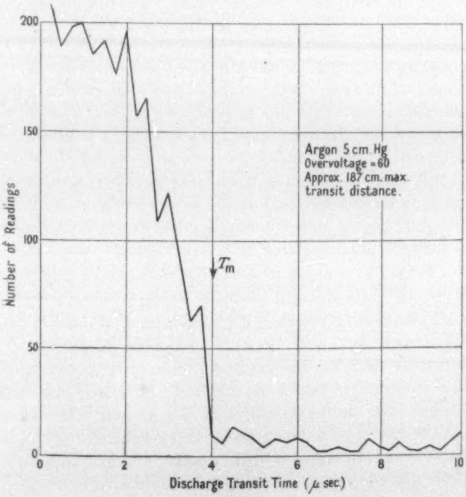

Figure 7.

indicating that statistical variations in the build-up time of the first avalanche (Wantuch 1947) were negligible. This experiment could be carried out with the 2 m. counter, mentioned in § 2, the cathode of which consisted of ten tubes 20 cm. long, when successive cylinders could be joined together in vaying numbers. The starting voltages (i.e. the counter voltage at which counts were first detected) for the various counter fillings are given in Table 2 for cylinder diameter 0·75 in. and wire diameter 0·005 in.

Table 2

Gas	Pressure (cm. Hg)	Starting voltage	Gas	Pressure (cm. Hg)	Starting voltage
Hydrogen	3	750	Argon	5	710
	5	920		10	920
	6	975		15	1140
	10	1240	Neon	10	400
	15	1530		20	440

Table 3

Argon pressure cm. Hg	Over-voltage		Time of propagation T (μsec.) and velocity of propagation V (cm/μsec.)				
			60·3 cm.	80·4 cm.	120·8 cm.	161 cm.	186·8 cm.
5	20	T	—	5·5	—	10·5	12·5
		V	—	14·6	—	15·3	14·9
		T	—	2·75	—	6·0	6·5
	40	V	—	29·4	—	26·8	28·7
		T	—	—	2·75	3·75	4·25
	60	V	—	—	44	42	44
10	20	T	—	7·25	—	13·5	—
		V	—	11·1	—	11·2	—
	40	T	—	3.5	—	7·5	—
		V	—	23	—	21·5	—
		T	—	2·5	—	5	—
	60	V	—	32	—	32	—
15	20	T	7	9·5	—	—	—
		V	8·6	8·6	—	—	—
	40	T	—	4·5	—	9	—
		V	—	17·9	—	17·9	—
		T	—	3	—	6·5	—
	60	V	—	27	—	25	—

Velocities of propagation, measured in argon at 5, 10 and 15 cm. Hg, for distances varying between 60·3 and 186·8 cm. are given in Table 3. It should be mentioned that these distances are arbitrarily measured between the mid-points of the start and stop cylinders, since the time to build up the pulse to an amplitude sufficient to trigger the timing circuit was not known. Errors arising in the resulting velocities could be reduced however by subtracting the observed time for propagation over a short cathode from that for a longer cathode. The results of Table 3 show that such errors are not noticeable.

§ 4. DISCUSSION OF RESULTS

The data plotted in Figures 4–6 represent the values of discharge propagation velocity, determined from the maximum transit time T_m as shown in Figure 7, and will, it is hoped, be of interest to those contemplating the use or study of externally quenched counters in circumstances where a knowledge of the time of discharge spread is of importance.

We had hoped that the results might have enabled a distinction to be made between the two mechanisms shown in Figure 1, but this hope has proved futile as the following discussion will show. The mechanism (*a*) of Figure 1 could, if effective, be related to photo-ionization processes (Alder *et al.* 1947, Wilkinson 1948), and its applicability to the case of self-quenched counters is strongly supported by the experiments of Stever (1942) and others. The mechanism (*b*) of Figure 1 has little fundamental interest and (Corson and Wilson 1948, Jaffe *et al.* 1949 and references there cited) is thought likely to apply in the case of externally quenched counters; although no proof of its existence has yet been found, the experiments with divided cylinder counters (Jaffe *et al.* 1949) certainly show that photons, capable of traversing some centimetres of counter length, and then producing either mechanism (*a*) or (*b*) of Figure 1, exist in large numbers in externally quenched counters. This fact does not, of course, show which mechanism is operative in causing discharge spread in a normal (continuous cathode) counter.

For mechanism (*b*) of Figure 1, the electron transit time across the counter is of the order of 10^{-7} second (Sherwin 1943, Corson and Wilson 1948, Curran and Craggs 1949), and probably most of this time will have passed, due to the electron movement in the low field region of the counter, before the photon necessary for spreading the discharge will have appeared. Therefore, the maximum velocity given in Figures 4–6, about 75 cm/μsec., will need axial photon movements of approximately 7·5 cm., and this quantity is reasonable (Jaffe *et al.* 1949). For mechanism (*a*) of Figure 1, elementary calculations show that if the operative photon is produced at a distance of a wire radius from the wire in a normal type of counter, a reasonable assumption in view of the Townsend avalanche building up near the wire, then it will be produced in about one three-thousandth of the full electron transit time of mechanism (*b*), approximately 10^{-10} second or less, which thus demands a photon step of about 0·0075 cm. for the maximum velocity of spread—or a shorter step for the lower velocities. Since the photo-ionizing radiations may have a value of μ (at 10 cm. Hg) of the order of 30 cm^{-1} (Jaffe *et al.* 1949, Loeb 1948) and are thus highly absorbable, giving 30% absorption for a path length of 0·1 mm., the photon step of approximately 0·075 mm. is not unreasonable and no decision between the two mechanisms of Figure 1 seems possible.

Further experiments are required, and a possible approach might be to prevent mechanism (*a*) from being operative by the use of many beads (Stever 1942, Wilkening and Kanne 1942) sealed on to the wire. It appears to be more difficult to avoid mechanism (*b*) although several possible methods have been considered. For example, it might appear attractive to consider the use of an axial magnetic field to turn back to the cathode the photo-electrons emitted there, but such a field has little or no effect since at Geiger counter pressures, the mean free path of the electron is too small to allow any appreciable deflection to occur (Lawson 1949). Consideration is also being given to possible experiments with cathodes consisting

of wires or bars (McCusker 1944) which should reduce cathode effects and with tubular cathodes of varying diameter, but both methods are difficult to arrange so as to give a clear cut differentiation between the two mechanisms.

ACKNOWLEDGMENTS

The authors wish to thank Sir Arthur Fleming, Director, and Mr. B. G. Churcher, Manager, of the Metropolitan-Vickers Research Department, for permission to publish this account of experiments carried out in their laboratories. The authors' thanks are also due to Mr. F. R. Perry (Metropolitan-Vickers, High Voltage Laboratory) for his support of the work, and to Professor J. M. Meek and Mr. W. Hopwood (Department of Electrical Engineering, University of Liverpool), and Dr. D. H. Wilkinson (Cavendish Laboratory) for discussions.

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Momenta in Atoms using the Thomas–Fermi Method

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MS. received 18*th August* 1949

ABSTRACT. The Thomas–Fermi statistical theory of the free atom is used to calculate the momentum distribution and the shape of the Compton profile for x-ray scattering, the results being expressed in dimensionless form applicable to all atoms. The mean momentum, which varies as $Z^{2/3}$, where Z is the atomic number, agrees well with more accurate wave–mechanical calculations. Numerical results are also reported for argon and krypton using the Dirac modification of the original theory, in which exchange effects are taken into account and the atom has a finite radius. Agreement with experiment is not noticeably better with these refinements.

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§ 1. INTRODUCTION

THE statistical theory of Thomas (1927) and Fermi (1928), to be referred to as T.F., has had many applications since its foundation. As a rough method of treating heavy atoms, for which wave-mechanical methods become impracticable, it has met with considerable success. Fermi in his original paper utilized the method to investigate angular momenta, and later Burkhardt (1936) showed how to obtain the linear momentum distribution and the Compton profile. We have carried out calculations on momentum distribution, mean momentum and the Compton profile, and the results are given here, in a dimensionless form applicable to all atoms. The significance of the shape of the Compton profile lies in the fact that it measures the distribution of electronic velocities in the atom, and is also closely related to the angular distribution of fast inelastically scattered electrons when a homogeneous beam impinges on the atom. The relation of all this to the momentum distribution has been studied recently by Duncanson and Coulson (1948).

Dirac (1930) has obtained a generalized Thomas–Fermi equation (T.F.D. equation) taking into account the exchange effect neglected in the simpler T.F. theory. In doing so, he has in fact shown that the T.F. method is really a semi-classical approximation to Hartree's self-consistent-field method; and the T.F.D. equation he obtains, by a similar approximation, from the Fock equations. Unfortunately, the model for a neutral atom given by the T.F.D. equation is not very satisfactory, since a discontinuity of charge density appears at the boundary of the atom. In spite of this, it seemed to us worth while to investigate whether the inclusion of the exchange term led to any improvements in the predictions concerning momenta. The only other work of this type seems to be some unpublished calculations of Konya, reported by Gombas (1948), taking account of exchange and correlation effects.

§2. MEAN RADIAL DISTRIBUTION FUNCTIONS FOR T.F. AND T.F.D. ATOMS

Below is a summary of the equations which we shall require. In this paper we restrict ourselves solely to the case of isolated neutral atoms. The notation for the T.F. equation is the same as that used by Slater and Krutter (1935). For the T.F.D. equation their ψ and d correspond to our ϕ and α.

T.F. method T.F.D. method

$$\frac{d^2\phi}{dx^2}=\frac{\phi^{3/2}}{x^{1/2}} \qquad\qquad \frac{d^2\phi}{dx^2}=x[\alpha+\{\phi/x\}^{1/2}]^3 \qquad \ldots\ldots(1)$$

with boundary conditions

$$\phi(0)=1, \quad \phi(\infty)=0 \qquad\qquad \left.\begin{aligned}\phi(0)=1, \quad \phi(x_0)&=\tfrac{1}{16}\alpha^2x_0\\ \phi'(x_0)&=\tfrac{1}{16}\alpha^2\end{aligned}\right\} \qquad \ldots\ldots(2)$$

$$\alpha=6^{1/3}/4(\pi Z)^{2/3} \qquad \ldots\ldots(3)$$

$$p_0(x)=\left(\frac{2\boldsymbol{me}^2Z}{b}\right)^{1/2}\left(\frac{\phi(x)}{x}\right)^{1/2} \qquad\qquad p_0(x)=\left(\frac{2\boldsymbol{me}^2Z}{b}\right)^{1/2}\left[\alpha+\left\{\frac{\phi}{x}\right\}^{1/2}\right] \qquad \ldots\ldots(4)$$

where x is a dimensionless variable defined in terms of the distance r from the nucleus by

$$r=bx=\left(\frac{3}{32\pi^2}\right)^{2/3}\frac{\boldsymbol{h}^2}{2\boldsymbol{me}^2Z^{1/3}}x, \qquad \ldots\ldots(5)$$

and bx_0 = radius of the atom in the T.F.D. method; $p_0(r)$ is the maximum momentum of an electron at a distance r from the nucleus and is related to the number of electrons per unit volume at r, $n(r)$ by

$$n(r)=\frac{8\pi}{3\boldsymbol{h}^3}p_0^{\,3}(r). \qquad \ldots\ldots(6)$$

Let us define $I_r(p)\,dp$ to be the probability of an electron at r having momentum whose absolute magnitude lies between p and $p+dp$. Then, since the density in momentum space is constant up to $p=p_0$,

$$\left.\begin{aligned}I_r(p)\,dp=\frac{4\pi p^2\,dp}{4\pi p_0^{\,3}/3}&=\frac{3p^2}{p_0^{\,3}}\,dp \quad \text{for } p\leqslant p_0\\ &=0 \qquad\quad\;\; \text{for } p>p_0.\end{aligned}\right\} \qquad \ldots\ldots(7)$$

If we draw the (r,p) plane for the T.F. atom, as shown in Figure 1, to each value r there corresponds a value $p_0(r)$, and a curve is defined dividing the (r,p) plane. $I_r(p)\,dp$ is different from zero only in the shaded region. Next, let us define $I(p)\,dp$ to be the probability of one electron having momentum between p and $p+dp$.

$$I(p)\,dp=\int_0^{r(p)}\frac{I_r(p)\,dp\,n(r)4\pi r^2\,dr}{Z}=\frac{32\pi^2}{3\boldsymbol{h}^3Z}r^3(p)\,p^2\,dp, \qquad \ldots\ldots(8)$$

where $r(p)$ is defined by equation (4) of the T.F. method. Equation (8) is the relation first derived by Burkhardt.

The modification for the T.F.D. atom is as follows. The region of the (r,p) plane over which $I_r(p)\,dp$ is different from zero is the shaded portion shown in Figure 2. The distance R defines the boundary of the atom on this model, the charge density being zero outside this distance. In this case

$$I(p)\,dp = \frac{32\pi^2}{3\boldsymbol{h}^3 Z} r^3(p) p^2\,dp \quad \text{for } p \geqslant p_0(R), \qquad \ldots\ldots(9)$$

$$= \frac{32\pi^2}{3\boldsymbol{h}^3 Z} R^3 p^2\,dp \quad \text{for } p \leqslant p_0(R), \qquad \ldots\ldots(10)$$

where $r(p)$ is now given by equation (4) of the T.F.D. method. From $I(p)$ the mean momentum

$$\bar{p} = \int_0^\infty p I(p)\,dp, \qquad \ldots\ldots(11)$$

and the Compton profile defined by (Duncanson and Coulson 1945)

$$J(q) = \frac{1}{2}\int_q^\infty \frac{I(p)}{p}\,dp, \qquad \ldots\ldots(12)$$

can be obtained.

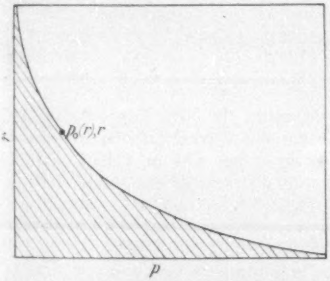

Figure 1. The (r, p) plane for T.F. atom.

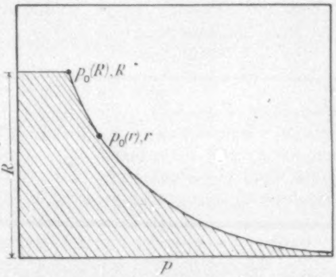

Figure 2. The (r, p) plane for the T.F.D. atom.

For the T.F. method it is convenient to introduce new quantities P, $I_0(P)$, $J_0(Q)$ defined by

$$P = \{\phi(x)/x\}^{1/2}, \qquad I_0(P)\,dP = I(p)\,dp, \qquad J_0(Q)\,dQ = J(q)\,dq. \qquad \ldots\ldots(13)$$

From these it follows that

$$I_0(P) = x^2 \phi(x) \qquad \ldots\ldots(14)$$

and

$$J_0(Q) = \frac{1}{2}\int_Q^\infty \frac{I_0(P)}{P}\,dP. \qquad \ldots\ldots(15)$$

In this way the calculations can be made in a form applicable to all atoms, since P, $I_0(P)$ and $J_0(Q)$ depend only on ϕ and x, which are dimensionless and independent of Z. For the T.F.D. method, however, since equation (1) has to be solved separately for each Z, calculations have to be made individually for each atom.

§ 3. SOLUTIONS USED

For the T.F. method our calculations are made for two solutions of the fundamental equation (1).

First, the approximation obtained by Sommerfeld (1932) is used. This has the form

$$\phi(x)=\{1+(x/a)^d\}^{-c} \qquad \ldots\ldots(16)$$

with $a=12^{2/3}$, $d=0{\cdot}772$, $c=3{\cdot}886$.

Secondly, we use the numerical solution of Bush and Caldwell (1931) together with an expansion for small x, due to Baker (1930), and an expansion which we have obtained for large x. This is of the form

$$\phi(x)=\frac{144}{x^3}\left\{1-\frac{F_1}{x^c}+\frac{F_2}{x^{2c}}-\frac{F_3}{x^{3c}}+\ldots\ldots+\frac{F_{10}}{x^{10c}}-\ldots\ldots\right\}, \qquad \ldots\ldots(17)$$

where $c=0{\cdot}772$.

All the coefficients in (17) may be obtained in terms of F_1, which is itself determined by fitting our expansion to the results of Bush and Caldwell. Coefficients are given in terms of F_1 in Table 1, and the best value we can obtain by this method is $F_1=13{\cdot}21$. For the T.F.D. equation the solutions obtained by Jensen, Meyer Gossler, and Rohde (1938) are used; the calculations are carried out for Ar and Kr. For Ar the solution used is one with an initial slope of $-1{\cdot}635403$ and a value of $x_0=12{\cdot}7$ (radius 2·27 A.); for Kr the initial slope is $-1{\cdot}6178552$ and $x_0=16{\cdot}7$ (radius 2·37 A.).

Table 1.

Coefficient	Value	Coefficient	Value	Coefficient	Value
F_2	$0{\cdot}62546F_1^2$	F_5	$0{\cdot}05507F_1^5$	F_8	$0{\cdot}00255F_1^8$
F_3	$0{\cdot}31340F_1^3$	F_6	$0{\cdot}02070F_1^6$	F_9	$0{\cdot}00085F_1^9$
F_4	$0{\cdot}13737F_1^4$	F_7	$0{\cdot}00741F_1^7$	F_{10}	$0{\cdot}00030F_1^{10}$

§ 4. RESULTS

For the Sommerfeld approximation some results can be obtained analytically. However, the following point arises here: the expression (8) is normalized such that

$$\int_0^\infty I(p)\,dp=\int_0^\infty I_0(P)\,dP=1$$

if $r(p)$ is given in terms of an exact solution ϕ of the original differential equation (1). The Sommerfeld expression is certainly not an exact solution, and for this we find that

$$\int_0^\infty I_0(P)\,dP=4c\Gamma\left(\frac{c}{2}\right)\Gamma(c)\Big/\Gamma\left(\frac{3c}{2}\right)^*, \qquad \ldots\ldots(18)$$

which equals 0·88 when $c=3{\cdot}886$.

For that reason we define

$$\overline{P}=\int_0^\infty PI_0(P)\,dP\Big/\int_0^\infty I_0(P)\,dP. \qquad \ldots\ldots(19)$$

Also we use this definition in the cases of Ar and Kr by the T.F.D. method, though in these cases the normalization factor is only slightly different from unity.

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* One of us (N.H.M.) has investigated this relation, when both sides of (18) are normalized to unity, as a method of obtaining a better approximation than Sommerfeld's over the whole range of x. The results will be published in a note elsewhere.

Using (19), we obtain for the Sommerfeld solution

$$\overline{P}=\frac{12^{2/3}}{16}\,\frac{\Gamma(5c/3)\Gamma(c/3)\Gamma(3c/2)}{\Gamma(2c)\Gamma(c)\Gamma(c/2)}=0{\cdot}449. \qquad \ldots\ldots(20)$$

From (4) and (13)

$$\bar{p}=2Z^{2/3}\left(\frac{4}{3\pi}\right)^{1/3}\overline{P}=0{\cdot}675Z^{2/3}\,\text{A.U.} \qquad \ldots\ldots(21)$$

If we retain (15) as defining $J_0(Q)$, then

$$J_0(0)=3\times 12^{1/3}c\Gamma(2c/3)\Gamma(c/3)/\Gamma(c)=6{\cdot}53. \qquad \ldots\ldots(22)$$

Figure 3, curve I, shows $I_0(P)$ against P for the Sommerfeld solution, whilst Figure 4, curve I, shows $J_0(Q)$ against Q.

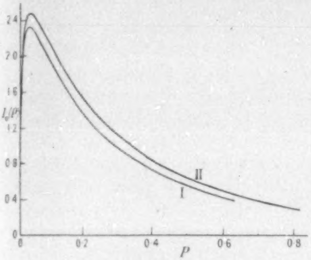

Figure 3. Momentum distribution function $I_0(P)$ for T.F. atom.

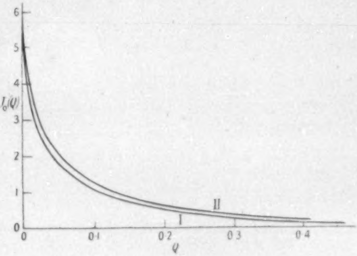

Figure 4. Shape of the Compton profile for T.F. atom.

For the second solution all the quantities have to be obtained numerically, and Figure 3, curve II, shows $I_0(P)$ against P, Figure 4, curve II, shows $J_0(Q)$ against Q. For this solution

$$\bar{p}=0{\cdot}688Z^{2/3}\,\text{A.U.} \qquad \ldots\ldots(23)$$

For both solutions the behaviour of $J_0(Q)$ for small Q can be obtained as follows. For small P,

$$I_0(P)\sim x^2\times 1/x^3=1/x, \qquad \ldots\ldots(24)$$

$$\mathrm{P}\sim 1/x^2, \qquad \ldots\ldots(25)$$

so that

$$J_0(Q)=J_0(0)-\frac{1}{2}\int_0^Q\frac{I_0(P)}{P}\,dP=J_0(0)-\text{const.}\times Q^{1/2}. \qquad \ldots\ldots(26)$$

Thus the slope is infinite at $Q=0$; this can be seen from Figure 4.

The calculations for the T.F.D. method have been carried out for Ar and Kr, the results being shown in Figures 5 and 6. Curves I correspond to Ar, curves II to Kr. Using a definition similar to (19) for Ar, $\bar{p}=4{\cdot}65$ A.U., for Kr, $\bar{p}=7{\cdot}27$ A.U.

For this case the behaviour of $J(q)$ for small q can also be obtained.

$$J(q)=J(0)-\frac{1}{2}\int_0^q\frac{I(p)}{p}\,dp. \qquad \ldots\ldots(27)$$

From (10), for small p,

$$I(p)\sim p^2, \qquad \ldots\ldots(28)$$

$$J(q)=J(0)-\text{const.}\times q^2. \qquad \ldots\ldots(29)$$

This gives $J(q)$ as shown in Figure 6.

§ 5. DISCUSSION OF RESULTS

Before discussing these results we should note that since the T.F. method is a statistical method, any periodic properties of atoms will not be found by this treatment. Now Duncanson and Coulson (1948) have shown that according to wave mechanics the mean momentum varies fairly smoothly with Z, so that we might expect the T.F. theory to give reasonable results for this quantity for heavy atoms. However, Duncanson and Coulson show further that the half-widths of the Compton profiles for various atoms have a decidedly periodic variation with atomic number, and therefore we can expect little agreement here, except possibly for the rare gases.

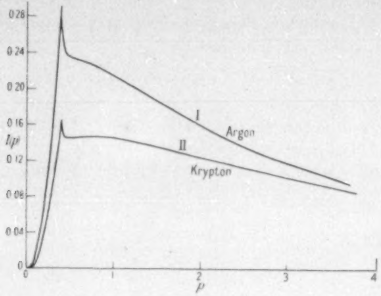

Figure 5. Momentum distribution function $I(p)$ for T.F.D. atom.

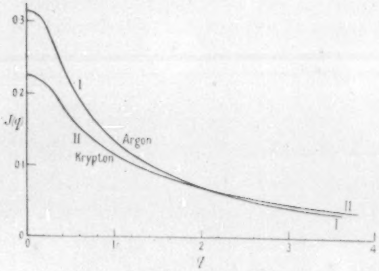

Figure 6. Shape of the Compton profile for T.F.D. atom.

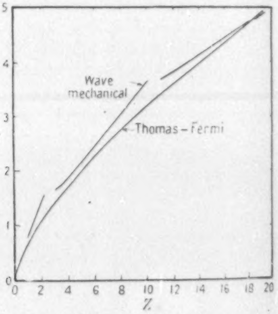

Figure 7. Comparison of mean momentum in T.F. atom and wave-mechanical atom.

When we compare the mean momentum given by the T.F. method with the wave-mechanical results of Duncanson and Coulson, we do find fairly good agreement. Figure 7 shows the results, plotted together with the wave-mechanical results. It is seen that the two sets of results approach more closely as the atomic number increases, a result to be expected.

With the T.F.D. method, the calculation has only been carried out for two atoms, so that, of course, no law of variation can be found. The numerical values of the mean momentum are slightly less than those given by the T.F. method.

However, when we turn to the Compton profile, we see immediately from the T.F. results in Figure 4 that the shape is wrong, the curve showing a cusp at $Q=0$. With a curve of this shape, the width at half-maximum is obviously no criterion by which to define the curve. A better criterion might be the width at half-area; we give this width in Table 2, realizing though that comparison with wave-mechanical results is not very significant. The explanation of the cusp seems to be that the majority of electrons with small momentum lie in the outer regions of the atom, where the electron density is low, and the assumptions of the theory therefore not valid. In fact, because of this, the ϕ function, from which all quantities are calculated, falls away much too slowly at large distances. An improvement will result if the method used cuts the atom off after a finite distance. This the T.F.D. method does, and for small q it gives the correct shape for the Compton profile. Table 3 gives the half-widths for Ar and Kr, and shows, unfortunately, that for Ar the quantitative comparison with the wave-mechanical value is not particularly good.

Table 2. Widths of Compton Profile at Half-Area by T.F. Method

Quantity	Sommerfeld	Full solution
Width in dimensionless units	0·214	0·216
Width in A.U.	$0{\cdot}322Z^{2/3}$	$0{\cdot}325Z^{2/3}$

Table 3. Widths of Compton Profile at Half-Maximum by T.F.D. Method

Element	Wave-mechnical	T.F.D.
Ar	2·24 A.U.	1·62 A.U.
Kr	—	2·08 A.U.

It should be mentioned also that the T.F.D. method inevitably gives a non-smooth curve for the momentum distribution (see Figure 5). This is caused by the discontinuity of charge density at the boundary. The cusp in Figure 5 will appear at the same value of p for all atoms. That this is so follows from the fact that the cusp always occurs at $p=p_0(R)$.

From (4) of T.F.D. method

$$p_0(x)=\frac{1}{\pi\alpha}[\alpha+\{\phi(x)/x\}^{1/2}].$$

At the boundary of the atom, $x=x_0$, and from (2), $\phi(x_0)/x_0=\frac{1}{16}\alpha^2$. Therefore

$$p_0(x_0)=5/4\pi\text{ A.U.}=0{\cdot}398\text{ A.U.}$$

The cusp will presumably become less pronounced as the atomic number increases (see Figure 5). On the whole it seems more satisfactory to have the Compton profile of the correct form rather than the momentum distribution, as it is this profile which is observed experimentally, and the momentum distribution inferred from it.

In conclusion, the method does not seem a very satisfactory one for momentum predictions, presumably because an important part of the momentum range is that of small momenta. The outer electrons provide the main contribution in this region, and for these electrons the fundamental assumptions of the theory are not really valid. Also for heavy atoms the relativistic correction should be considered; but the relativistic T.F. equation, as formulated at present, is very unsatisfactory.

ACKNOWLEDGMENT

One of us (N.H.M.) would like to acknowledge a grant from the Department of Scientific and Industrial Research.

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A Quantitative Study of the Domain Structure of Single Crystals of Silicon-Iron by the Powder Pattern Technique

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MS. received 5th October 1949

ABSTRACT. A quantitative study of the domain structure of single crystals of silicon–iron has been made by the powder pattern method, special examination being made of the patterns formed on a strip specimen whose surface was approximately a (100) plane with a [011] direction parallel to the long edge of the strip. For Mode III magnetization, which occurs when a field acts inside the specimen parallel to the [011] direction, the line deposits formed at right angles thereto were on the average spaced in good accord with Néel's theory in the case of wide strips, but in less good accord for narrow strips. Qualitative results of other measurements on strip and disc specimens are reported and interpreted.

§ 1. INTRODUCTION

It is well known that in order to explain the fact that a piece of ferromagnetic material may exist in an unmagnetized state and yet may be magnetized to technical saturation in a field of a few oersteds, Weiss introduced the concept of the ferromagnetic domain. A ferromagnetic crystal consists of a large number of small regions or domains each magnetized to an intensity of spontaneous magnetization, I_s, appropriate to the temperature, along one of the easy directions of magnetization in the crystal. In the unmagnetized state the distribution of the magnetic vectors of these regions is such that the specimen does not exhibit bulk magnetization. In recent years the domain concept has been subjected to much critical examination. In particular, much attention has been paid (Néel 1944) to the grave difficulties which must arise with free magnetic charges unless it is postulated that the normal component of the magnetization across a boundary or Bloch wall between two adjacent domains is continuous. It has also been found necessary to postulate that such domain boundaries must be perpendicular to any effective magnetic field which acts inside a single crystal, as described below. In applying these postulates to any particular crystal specimen, the shape of the specimen is important and simple expressions for longitudinal and transverse magnetizations have been obtained in three cases only (Néel 1944, Lawton and Stewart 1948, Lawton 1949), namely, that of a specimen in the form of an oblate spheroid whose equatorial plane is a (100) plane parallel to which a field is applied, that of a long rod or ellipsoidal specimen, in which case the transverse magnetization under any applied field is bound to be small, and, finally, that of an oblate spheroid whose equatorial plane is a (110) plane parallel to which a field is applied.

It is only necessary to give here a brief outline of what may be presumed to happen in the second case. If we consider an iron crystal, a common-sense argument leads us to conclude that under an infinitely small effective field H each of the domain vectors will be turned into one of the three directions of easy magnetization favoured by this field; or, using Néel's terminology, we may say that the six-phase state or Mode I magnetization gives place to a three-phase state or Mode II magnetization. It must be emphasized that H is not an external field but the field acting *inside* the crystal specimen. Now, in order to maintain this condition and to satisfy the above postulates, it follows that H must lie along a [111] direction, and that the domains must be leaf-shaped with their boundaries perpendicular thereto. As the external field applied to the ellipsoid is increased, H is also increased and, in due course, a two-phase state or Mode III magnetization is established, when H must lie along a [110] axis and the two sets of leaf-like domains have their boundaries perpendicular thereto. Finally, when we make the applied field very strong, all domain vectors will rotate to set parallel to the applied field, and we have a single-phase state or Mode IV magnetization; this may be particularly important with disc-shaped specimens.

A simple model may help to make these new ideas more clear. Let us slice a crystal cube of iron parallel to a (110) plane, and let us imagine that it is magnetized according to Mode II by an external field parallel to the surface of the slice. We should then obtain a set of domains with their boundaries perpendicular to the ternary axis, and the intersections of these boundaries on the (110) plane should form a series of lines perpendicular to the [111] direction; such lines —we will call them *ternary lines*—are found by the Bitter powder technique described below.

Let us now consider a slice cut parallel to the (100) face, and from it turn a disc on which a [110] axis may be marked. Applying a sufficient field parallel to the plane of the disc, not necessarily along the [110] direction, we should get a definite set of lines perpendicular to this [110] direction when Mode III magnetization is established. Such lines are found by experiment: we will call them *binary lines*.

In Figure 1 is depicted a special case of a long wide strip of a single iron crystal, cut as shown, which was studied in detail by Néel. The figure shows that the upper surface of the strip is a (100) plane and that the [011] direction lies in the middle of this surface. Parallel to the [011] direction is applied an effective field H; this is the field actually measured by a magnetic potentiometer (Bates 1945, 1948). When Mode III magnetization obtains, the main or primary domains form regular leaf-like or plate-like sections perpendicular to the [011] axis. Successively, each domain is magnetized parallel either to the direction labelled I_1 or to that labelled I_2. As the field strength increases beyond the small minimum value required for Mode III magnetization, I_1 and I_2 leave the directions of easy magnetization, for which $\theta = 45^\circ$, to approach symmetrically more and more closely to the [011] direction. The intersections of the main domain boundaries with the (100) surface must therefore be a regular series of parallel lines perpendicular to the [011] axis.

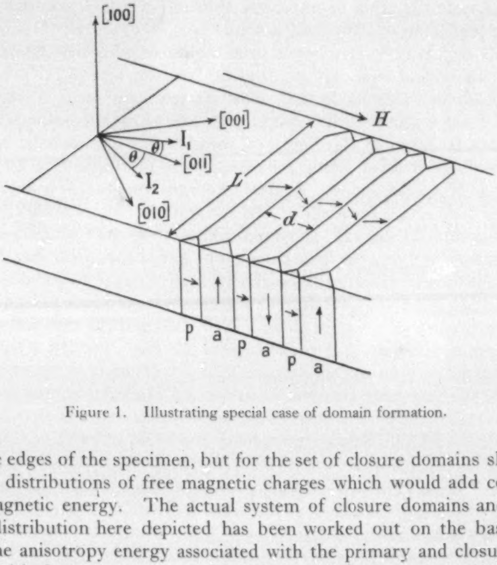

Figure 1. Illustrating special case of domain formation.

At the edges of the specimen, but for the set of closure domains shown there would be distributions of free magnetic charges which would add considerably to the magnetic energy. The actual system of closure domains and the main domain distribution here depicted has been worked out on the basis that the sum of the anisotropy energy associated with the primary and closure domains together with the energy stored in the Bloch walls or boundaries between them must be a minimum. On this basis the distance d between successive pairs of domain boundaries, which we may appropriately term the period of the primary domain structure, is given by $d = [\gamma L(W_\mathrm{a} + W_\mathrm{p})/W_\mathrm{a} W_\mathrm{p}]^{\frac{1}{2}}$, where γ is the energy per cm^2 of a domain boundary and can be found in terms of θ; W_a and W_p are the energies of formation per cm^3 of the two types, a and p, of closure domains, and can be expressed in terms of θ and the anisotropy constant K_1, (cf. Bates 1948); L is the width of the strip.

The present communication is an attempt at a quantitative experimental examination of the above results and, in particular, of the relation deduced for *d*. Short preliminary accounts of some of this work were given at Amsterdam in the summer of 1948 (Bates and Neale 1949) and in the course of the fourth Holweck lecture in Paris, last June (Bates 1949).

§ 2. EXPERIMENTAL PROCEDURE

The main method of studying domain structure is undoubtedly the Bitter figure or powder pattern method. This is essentially a kind of field-plotting on a miniature scale. The domain structures give rise to periodic variations in the emergent fields just above the surface of a ferromagnetic crystal, and regions where the stray field is most intense are located by allowing small magnetic dipoles to settle upon the surface from a colloidal or semi-colloidal solution. When the contrast between the metal surface and the deposited particles is adequate, the resulting figure or powder pattern can be photographed by means of a metallurgical microscope, good photographs being normally obtained with a magnification between 10 and 100.

We used an ordinary metallurgical microscope, with an internally reflecting prism to direct a beam of light from a 100-watt ' Pointolite ' lamp on to the surface of the crystal. The microscope was always used in the vertical position, and set for magnifications of 14 or 62 in most of the work. The microscope stage was removed from the microscope itself and mounted on a movable carriage about 20 cm. below its normal position, and the magnetizing systems were placed upon it. The specimen, in the form of a thin strip or disc, was placed with its face horizontal either across or in the gap of a small electromagnet giving a horizontal field, or upon the end face of an iron core standing in a vertical solenoid. In appropriate cases the effective field on the specimen was measured in a separate series of experiments with a small magnetic potentiometer. It is practically impossible, of course, to use the magnetic potentiometer when the surface of the specimen is covered with liquid during observation of the patterns. One can only replace the specimen between observations with great care in a standard position. With our arrangement we could detect fields of the order of 0·05 oersted, and measure fields of the order of 10 oersteds to an accuracy of 1%.

A drop of colloid solution was placed on the crystal surface and formed, when necessary, into a shallow cell of mean depth about 1 mm. by a tilted microscope cover slip placed over the drop. A tilted slip was necessary to avoid light reflected from the liquid surface.

Bitter (1932) originally used finely divided iron or γ-ferric oxide particles, suspended in alcohol or other liquids of low viscosity to enable them to settle quickly. We tried several original methods of deposition, but we did not succeed in surpassing the results obtained with colloidal magnetite prepared according to the Elmore–Lefort (Elmore 1938) recipe. The liquid thus prepared contains particles of all sizes, some forming a true colloid, some being merely in suspension, which settle on standing. After a short time the liquid forms a gel, but it can again be used after boiling it for a short time.

The preparation of the crystal surface is very important. Hand-polishing causes strains which result in variable and indeterminate domain structures; most of the beautiful ' maze ' patterns which have been published are merely records of surface strain. We advise polishing the surface with emery papers down to 000, giving a final polish by electrolysis, using Jacquet's solution (Merchant 1947), and later annealing the specimen *in vacuo*. During electrolytic polishing it is desirable to protect portions of the specimen which are not to be polished, and which would otherwise be eaten away, by coating them with polystyrene glue.

The axial orientations of the specimen were found by the maximum reflections from etch pits using a two-circle crystal goniometer. An independent check on the goniometer measurements was provided by measurements with a simple torque magnetometer (Webster 1924), in the case of disc specimens.

§ 3. GENERAL RESULTS WITH STRIP SPECIMENS

It is desirable to repeat that the above expression for d and the details of Figure 1 were calculated for pure unstrained iron and for a crystal with a (100) plane lying exactly in its upper surface. The crystals which we used were large single grains cut from a sheet of 2·8% silicon–iron, kindly supplied by Mr. G. Richer. Consequently, we could not expect exact numerical agreement with Néel's theory. In a very comprehensive treatment of powder patterns, Bozorth, Williams and Shockley (1949) have discussed the special features which arise, particularly in Mode II magnetization, when the crystal surface is not exactly coincident with a (100) plane etc. The main series of our experiments consisted in finding how the period d depended on the field H and on the width L of the specimen magnetized in Mode III.

We therefore started with a strip some 4·5 cm. long and of initial width L_1 about 2 cm. The strip was later cut to give different widths ranging from 2 to 0·4 cm. The surface of the specimen made an angle of not more than four degrees with the true (100) plane, but this might have been enough to produce a very shallow secondary domain structure on the surface or, alternatively, to give rise to a vertical component of magnetic field, so that, as explained later, successive line deposits were formed regularly at distances d, instead of $d/2$, apart. We did not try to make observations on the edges of the strip, as the secondary domain structure was not clear enough on our specimens.

The strip under investigation was magnetized by placing it in the horizontal field of an electromagnet, resting the ends of the strip on the flat sides of the pole pieces. The spacing d was measured by casting an image of the surface on to the ground glass camera screen and counting the number of lines in a 7 mm. section of the strip parallel to the field. The magnification was measured directly by using a fine, graduated scale as object. At moderate fields, this counting method was accurate to about 3%, the accuracy being limited by the shortness and indefiniteness of some of the lines. In any case the spacing varied over the strip, particularly at low fields, where the effects of pits and inclusions were important. But the quantity d is an *average* spacing value, and the above inaccuracy was countered by taking large numbers of readings. This was a long and tedious business, as after each measurement the strip had to be removed from the field and washed to remove colloid particles which had stuck to its surface. In addition, photographs were frequently taken and provided extra data.

The original strip L_1 was reduced in width to make strip L_2 by cutting a narrow strip from the former with a sharp razor. The two strips, L_2 and the fragment, were mounted side by side across the electromagnet gap to simulate as closely as possible the conditions under which L_1 was magnetized, so that the same field calibration might be used. Strip L_3 was similarly mounted, but with strip L_4 a new field calibration was made and used with strips L_5 and L_6 as well.

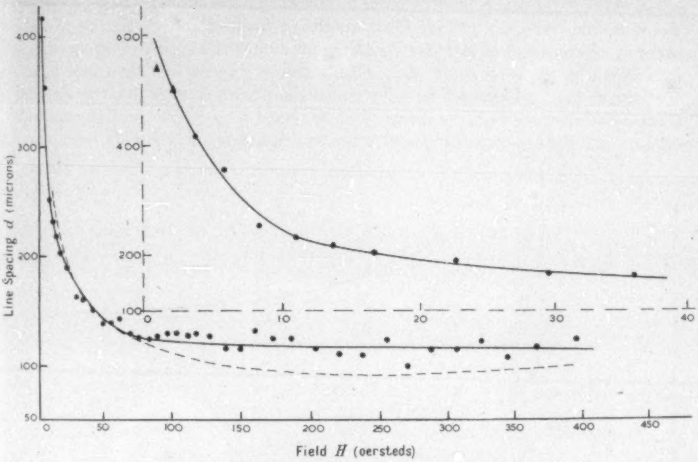

Figure 2. Domain spacing for strip width $L_1 = 1{\cdot}942$ cm.
—— Experimental curves. -------- M. Néel.

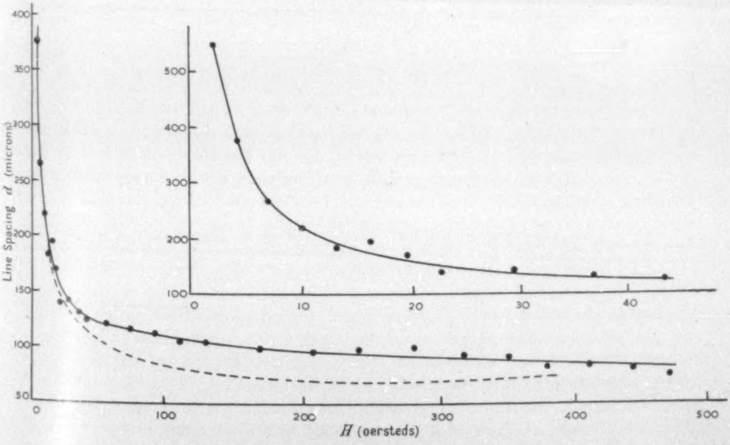

Figure 3. Domain spacing for strip width $L_4 = 1{\cdot}138$ cm.

The results are plotted graphically in Figures 2 to 5, and some important aspects of these graphs will now be considered. Firstly, with the data available, it was not possible to determine whether there are any real discontinuities in the (d, H) curves. Sixtus (1937) thought that there were discontinuities connected with the abrupt changes in the slope of the (I, H) curve, but his deductions appear to be based on a relatively small number of observations. Some apparent discontinuities are seen on individual graphs of Figures 2 to 4, but since these occur at different values of H for the different strips, the results are more fairly interpreted by the smooth curves. Néel's theory gives a smooth curve which is shown dotted in Figures 2 to 4 for comparison with our results. Individual points lie off the smooth curves because of the essentially discontinuous motion of domain boundaries accompanying domain multiplication as the field increases.

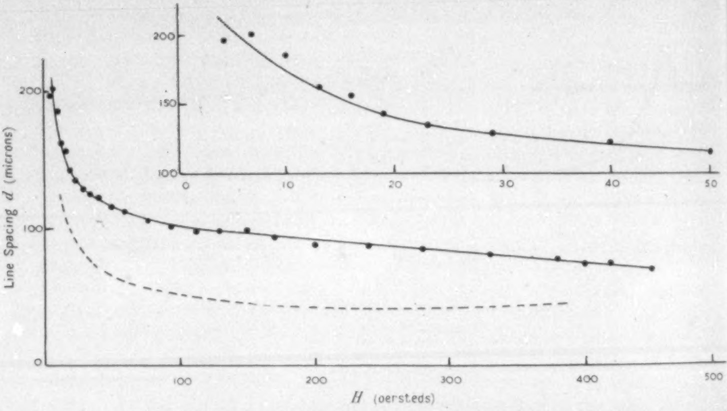

Figure 4. Domain spacing for strip width L_6=0·415 cm.

It was found experimentally that the points were the less scattered the larger the area of the strip for which the average spacing was measured; so that not only was the counting error reduced, but so also was the error introduced by the sudden movement of a domain boundary at the edge of the region measured, following a very small field increment. Thus the plotted experimental points are less scattered for the higher field values, where the average value of d is measured for a much larger number of domains than at low fields. The attainable accuracy is limited however by the area over which the field is uniform, i.e. the middle third of the portion of the strip between the pole pieces.

The theoretical curves in Figures 2 to 4 are calculated from Néel's data by multiplying his values of d for a strip 1 cm. wide by the appropriate value of $\sqrt{L}$. These curves are not, as explained above, the true theoretical curves for the silicon–iron material used here, since γ, W_a and W_p depend on the anisotropy constant K_1; K_1 for iron is usually taken as $4{\cdot}2\times10^5$ erg/cm^3, and $2{\cdot}8\times10^5$ for $3{\cdot}85\%$ silicon–iron. Now, calculation shows that d is proportional to $K_1^{-1/4}$ and to $I_s^{1/2}$, so that the effects of these differences in K_1 and those in I_s need not trouble us unduly. Néel's figures certainly give a curve closely approximating to the true theoretical curves for the different strip widths we used, and in the present state of our knowledge of K_1 in particular, more elaborate arithmetical calculation is unnecessary.

The agreement between the modified Néel curves and our results is good for wide strips, but not for narrow ones, the discrepancy being most marked for the narrow strips L_5 and L_6. This discrepancy is further brought out by the graphs of d against $\sqrt{L}$ for various field strengths in Figure 5. It is observed that the reduction in line spacing d on cutting down the strip is much less than would be expected. In fact, d depends upon L to a much smaller extent than is suggested by writing $d \propto \sqrt{L}$. Except for L_5 and L_6 the experimental values of d are below the theoretical curve at the lower field values and above it at higher fields. The maximum difference for the larger strip widths, about 20%, is still not great, considering the rapid variation of d with H at low field values. A notable

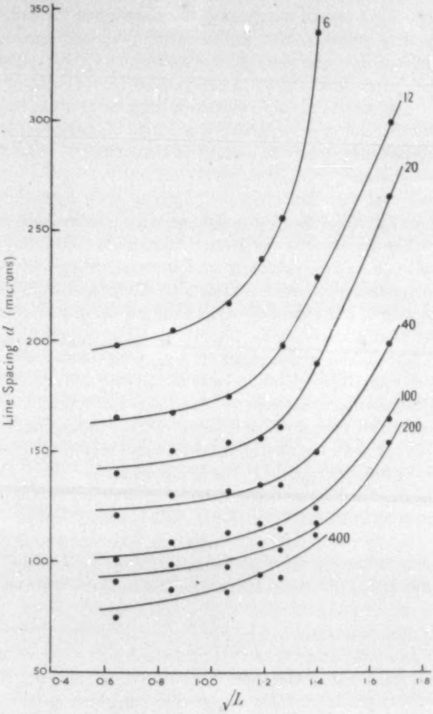

Figure 5. Variation of spacing of powder deposits as a function of strip width for field values stated in oersteds.

difference between the two, however, is the absence of any minimum value of d in the experimental curves. In all cases, d was found to decrease continuously as H increased until, at saturation, the regular domain boundaries disappeared and the deposits formed irregular streaks perpendicular to the field.

§4. RESULTS FOR INDIVIDUAL STRIP SPECIMENS

Strip Width L_1. Attempts to obtain patterns on the strip exactly as cut from the original unpolished crystal were unsuccessful owing to the irregularities in the surface produced by rolling. After polishing the strip with grade 2 emery paper the magnetization curve was determined and it was then possible to obtain regular boundary deposits, as shown in Figure 6(*a*), (*b*) for fields of 33 and 193 oe. respectively.* Not all of the patterns were as regular as those reproduced here, but they showed that this type of pattern was due to the internal domain structure of the strip and not due merely to a superficial structure. The deep scratches, clearly visible in Figure 6, would completely destroy any regular surface structure. Measurements of the variation of deposit spacing with field were taken as the strip was reduced in thickness by polishing with successively finer grades of emery paper. The results were not accurate enough to determine whether there was any variation in d with thickness of the strip, though the graphs for the initial stages of polishing showed a somewhat ill-defined minimum value of d at about 200 oe. which was absent after the final polishing and in narrower strips.

The patterns were not very regular after the final electropolishing, but improved considerably after annealing the strip. Thus, while strain did not affect the average domain width very much, it was found to produce irregular domain boundary shapes and shorter deposits. The curves in Figure 2 are for the electropolished and annealed specimen, and some corresponding patterns are shown in Figure 7. These were not all taken in the same region of the strip, but they show how the spacing decreases as the field increases.

Strip Width L_2. Owing to slight deterioration of the surface, after cutting down the original strip it had to be re-polished electrolytically. The thickness was only very slightly reduced by this operation, so it was assumed that the field acting on the strip L_2 was the same as that acting on L_1 for the same electromagnet current. The patterns for this width are shown in Figure 8 at the stated magnification. Measurements for the graphs were always made on the lower magnification patterns.

With strip L_2 it was discovered that patterns much more regular than those previously observed could be obtained, if, instead of simply reversing the magnet current many times to obtain the cyclic state corresponding to a given point on the (I, H) curve, the current was finally switched on and off several times. Thus, before placing the colloid on the strip, the following procedure was carried out to obtain a given state of magnetization: A, the electromagnet current was set at a given value either by starting from the demagnetized state or by increasing from a lower current; B, the current was reversed many times at this value, thus traversing the cycle between the points H, I and $-H$, $-I$; C, the current was switched on and off many times, so traversing the secondary loop between the points H, I and $H=0$, $I=I_{\text{rem}}$. Photographs (*d*) and (*c*) of Figure 8 show the same region at the same field, (*d*) being taken at the end of the operation B and (*c*) following operation C; they show a striking improvement in regularity of the lines. Later photographs and the points on the (d, H) graphs for the strip widths L_2 to L_6 were all taken using the new procedure.

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* For Figures 6–14 see Plates.

With L_2, patterns were also obtained on magnetizing it perpendicular to the long edges, i.e. in the $[01\bar{1}]$ direction instead of the $[011]$ direction. The ends of the strip were irregular in shape, making it difficult to assign an accurate value to the width L for this case; but a (d, H) graph was plotted over a short range of field values to provide additional points for insertion on the $(d, \sqrt{L})$ graph in Figure 5.

Strip Width L_3. No photographs were taken for this strip width.

Strip Width L_4. Two (d, H) graphs were plotted for this case, though only one is shown (Figure 3). The first was obtained using the three narrow strips cut off and placed alongside L_4, using the same field calibration as for L_1. Then the (I, H) curve and the field–current calibration curve were redetermined using L_4 alone, after which a second (d, H) graph, Figure 3, was obtained. There are considerable discrepancies between the two graphs at low field values, probably due to the greater inaccuracy in the determination of the field for the narrower strip. For the $(d, \sqrt{L})$ graph, of Figure 5, the points of width L_4 (up to 20 oe.) were taken from the first of these (d, H) graphs; beyond 20 oe. they were taken from the second graph.

Figure 9 shows some unusual and some extremely regular patterns on this strip; (*a*) and (*b*) are peculiar in that two of the boundaries carry heavy deposits for part of their length while the rest of the boundary is indicated only by a fine deposit. No perceptible reason for this (e.g. a hole or a pit at the boundary) was found on washing off the deposit. Photographs were also taken to show patterns on the whole strip and part of the pole pieces at a much lower magnification; the lines at the higher field were then only just visible. At about 6 oe., for example, they showed large variations in d over the specimen. The lines were more closely spaced, at all fields, near the pole pieces than at the centre, indicating, of course, that the field was greater there—an observation confirmed, as one would expect, by direct field measurement with the magnetic potentiometer.

Strip Widths L_5 and L_6. After cutting L_4 into two unequal parts the patterns obtained on the two strips were very irregular in comparison with those for wider strips. Measurements were taken after re-annealing and electropolishing the strips, using them side by side in the magnet field and taking measurements and photographs simultaneously (cf. Figure 4).

§ 5. A DETAILED STUDY OF DOMAIN BOUNDARY MOVEMENTS

A study of domain boundary changes consequent on field changes was made primarily to find out whether or not the deposits showed the same kind of shifting effect as has often been observed by earlier workers with deposits on reversing a *vertical field* acting on the specimen. The preceding work showed that the deposit spacing was equal to the width of a domain-pair d. For example, Figure 8(*a*) shows deposits with streaks on either side running at approximately $\pm 45°$ to the direction of the main line deposits. This is because the domain magnetizations on either side of a line deposit are at nearly $+45°$ and $-45°$ to the field direction. The patterns show that the domain magnetization changes in direction by 90° between one deposit and the next, so we conclude that there is here an intermediate boundary not revealed by any deposit. This conclusion was confirmed by a study of the deposits under changing field conditions.

Figure 10 shows the changes which occur on reversing and increasing the field. In (*a*) the pattern was produced by reversing a field of 17 oe. many times, and then traversing the region of the (I, H) curve between the remanence point and the point corresponding to +17 oe. In (*b*) the deposits have all shifted after a single reversal to $H = -17$ oe. On reversing again to +17 oe. all but one of the deposits shifted back to their original positions as shown in (*c*). These processes were also found to occur in smaller fields as well, most of the deposits shifting on reversing the field. Figure 10 (*d*) and (*e*), shows patterns in the same region produced by simple increases in the field to +41 oe. and to +190 oe. respectively, without reversing it. In the whole range from 17 oe. to 190 oe. the boundaries moved discontinuously. They also multiplied by the splitting of single domains, when a simple increase in the field produced unevenly spaced lines (an unstable state), as shown in (*e*). On switching the current on and off several times at the new value the deposits moved laterally again so as to become more evenly spaced, as shown in (*f*). Reversing the field to −190 oe. did not produce such a marked change in the pattern as occurred at lower fields, and only a few of the deposits moved to the positions shown in (*g*).

At near-saturation fields the deposits broadened considerably, and the streaks made a smaller angle with the deposits, consistent with the decrease in θ, the angle between a domain magnetization vector and the field. The deposits became narrower and straighter but retained the same average spacing as the magnetization was reduced to the remanent value. It then required only a small field in the reverse direction to cause the remanence pattern to break up completely; this occurred suddenly as the field increased through about 4 oe. in the reverse direction, i.e. on the steep part of the (I, H) loop below the coercive point.

§ 6. PATTERNS ON A STRIP IN DEMAGNETIZED STATE AND IN VERTICAL FIELDS

The strip was carefully demagnetized by reversal of a slowly decreasing field applied parallel to its surface. On the wide strip L_1, patterns not shown in this paper were obtained after the strip was electropolished but before it was annealed. They consist of bands running in the [010] and [001] directions, together with a small scale pattern of fine lines within the streaks in the same directions. The form of the patterns was the same all over the strip, but the band spacing varied. Similar bands were observed on the strip during the mechanical polishing, indicating the presence of a deep seated structure of domains with their boundaries parallel to (010) and (001) planes. The fine lines observed after electropolishing show the presence of a secondary structure due to surface strain. After annealing, these lines were absent, as shown in Figure 11, which reproduces the patterns observed with strip L_2. The principal structure still consists of domains magnetized in the [010] and [001] directions, but there are in addition several types of well defined, tapered closure domains, similar to those observed by Bozorth, Williams and Shockley (1949).

Where the surface was more steeply inclined to the (100) plane the closure domains covered a greater area of the surface, and between them heavier deposits of the arrowhead type were formed, in regions where the flux from the underlying domains formed dense, free surface poles. The shifting effect, similar to that observed with the patterns of Figure 12 which were obtained on a disc as described below, was also obtained with these deposits on reversing a field perpendicular to the strip surface.

Patterns on strip magnetized parallel to [011] axis
Direction of [011]→ in all cases

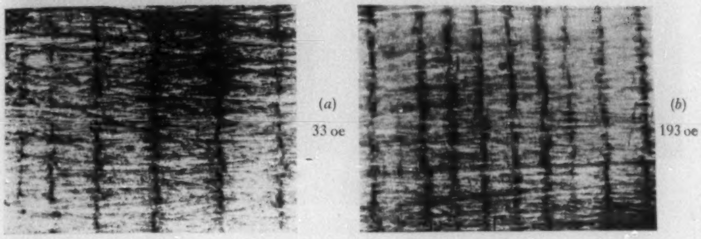

(a)
33 oe
(b)
193 oe

Figure 6. Strip width L_1. Patterns after mechanical polishing.

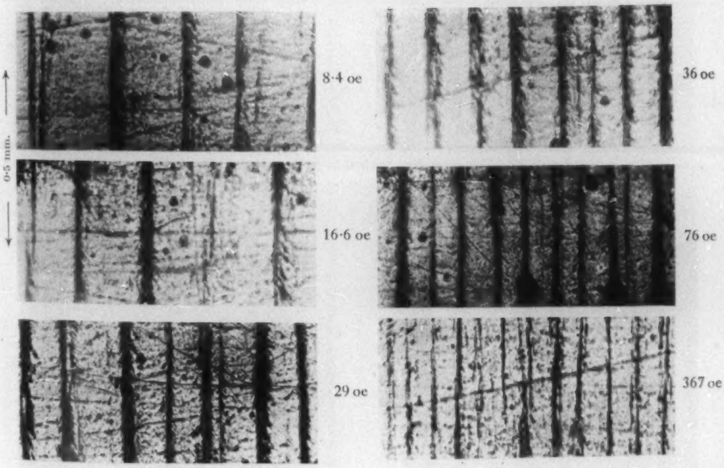
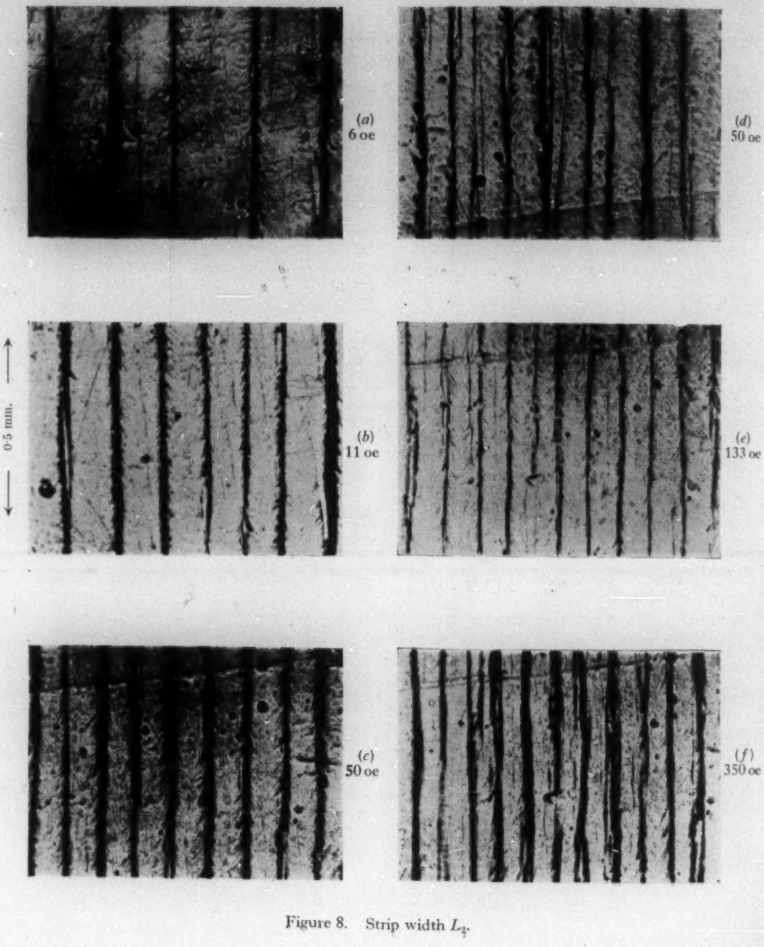

0·5 mm.
8·4 oe
16·6 oe
29 oe
36 oe
76 oe
367 oe

Figure 7. Strip width L_1. Patterns after electropolishing.



(*a*) 6 oe

(*d*) 50 oe

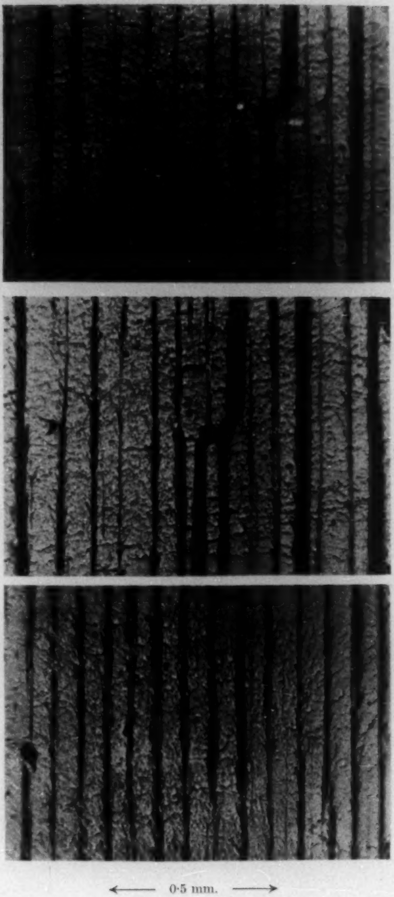
(*b*) 11 oe

(*e*) 133 oe

(*c*) 50 oe

(*f*) 350 oe

Figure 8. Strip width L_2.

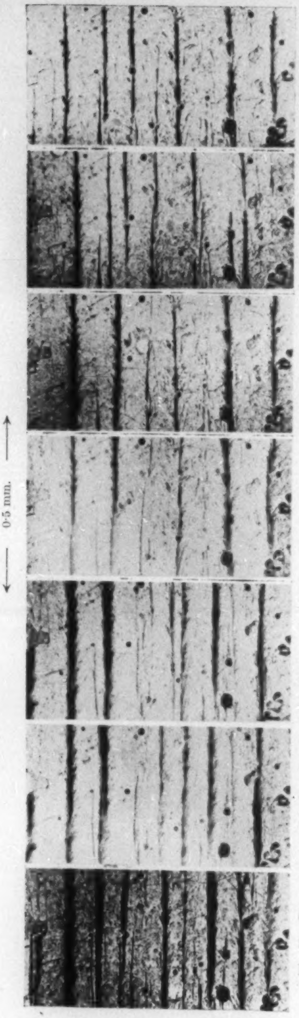


(*a*) 169 oe

(*b*) 207 oe

(*c*) 350 oe

Figure 9.



(*a*)
H = 17 oe→

(*b*)
Single reversal
H = 17 oe←

(*c*)
Single reversal
H = 17 oe→

(*d*)
Field increased slowly without reversing
H = 41 oe→

(*e*)
Further slow increase in field
H = 190 oe→

(*f*)
Field varied between remanence point and +190 oe
H = 190 oe→

(*g*)
Single reversal
H = 190 oe←

Figure 10. Patterns on strip L_4 in same region for varying fields.

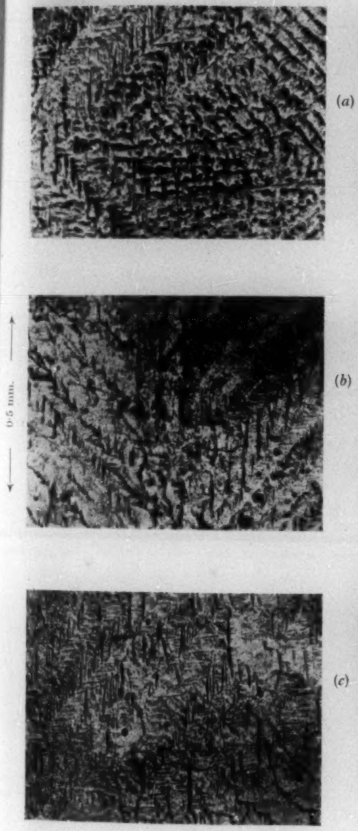


Figure 11. Patterns in demagnetized state, obtained on strip L_2 after annealing. $[01\bar{1}]$ axis →

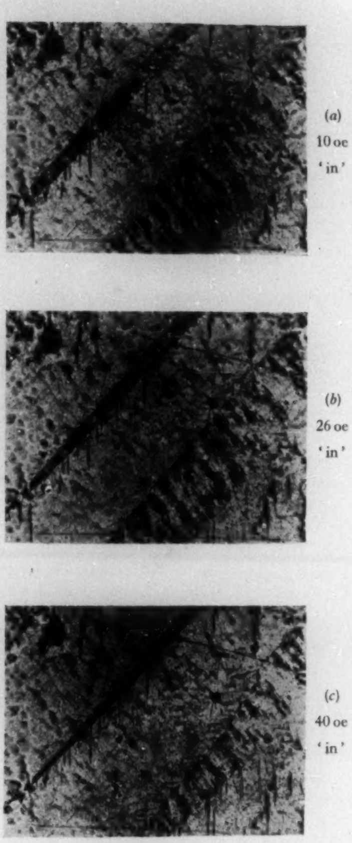
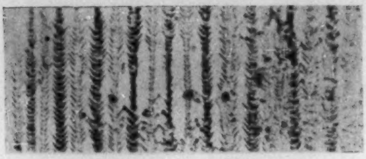
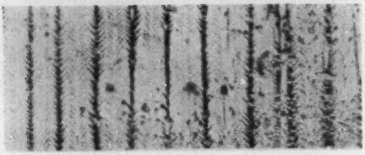

(a)
10 oe
'in'
(b)
26 oe
'in'
(c)
40 oe
'in'

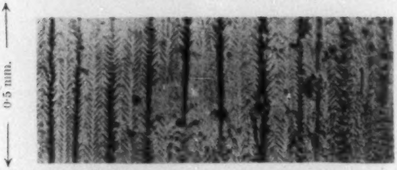
Figure 13. Patterns obtained on strip L_2 in a varying field applied perpendicular to surface, showing unusual moving boundary effect. $[01\bar{1}]$ axis→



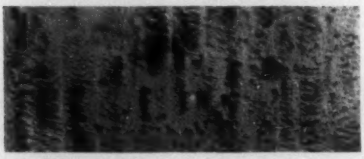
(*a*) H=0, d=56 μ



(*b*) H=120 oe, 'in'

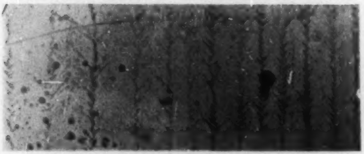


(*c*) H=120 oe, 'out'



(*d*) H=800 oe, 'out'

Figure 12 Patterns on single crystal disc in demagnetized state and in vertical field.



H=8 oe ↖

Figure 14. Pattern on disc as Figure 12, but in horizontal field, showing variation in line spacing and shifting effect as in vertical field.

An unusual moving boundary effect occurred with another type of pattern on varying the magnitude of a vertical field. Figure 13 shows a region of the surface almost exactly parallel to the (100) plane. There are prominent domain boundaries parallel to the (001) plane and very few closure domains. The underlying domains are magnetized parallel to the [010] and [0$\bar{1}$0] directions as shown by the streaky deposits perpendicular to these directions. But alternate domains, magnetized in one of these directions, increased in size at the expense of their neighbours on increasing a vertical field. Figures 13(*a*), (*b*) and (*c*) show this effect with vertical fields of 10, 26 and 40 oe. respectively. The vertical applied field, measured by a flat search coil in contact with the specimen, increased the vertical component of magnetization of those domains which decreased in volume, and this is confirmed by the increasing density of deposit on these domains (black in the photographs) as H increases. The alternate set of domains originally had a vertical component of magnetization in a direction anti-parallel to the field, and this is decreased on increasing the field; the horizontal component is correspondingly increased and the domains also increase in volume. Thus the net effect is to increase the total magnetization parallel to the field. As with a horizontal field, the domain boundary movements took place discontinuously. They were also irreversible, except for small field changes, and never reformed in the same positions on repeated application of the same field.

§ 7. EXPERIMENTS WITH DISC SPECIMENS

It would seem at first sight that it would be possible to take a disc, cut from a sheet specimen with a (100) plane in its surface, apply a field parallel to its surface and along the [011] direction, and so produce a series of domain boundaries perpendicular to the latter direction. The boundaries should then be wide apart in the middle of the disc and should close up as one moves out from the centre along the [011] direction; and it might be thought possible to verify the Néel expression for d, and in particular its dependence on L, in a very simple direct manner with one disc. However, discs have to be cut and it is difficult to cut them without causing distortions etc., and the effects of pits and inclusions are much greater than with strip specimens. Hence, we have so far only made qualitative measurements with discs. Some of these will now be described.

Disc No. 1. The first disc, of diameter 1·28 cm., was cut from a 2·8% silicon–iron crystal whose face was practically a (100) plane, the [100] direction making an angle of less than 3° with the surface normal. It was polished in orthophosphoric acid (before it was decided that Jacquet's solution was best for our purposes) which left it rather thin (0·173 mm.), rather badly pitted, but reasonably free from strain. Figure 12 (*a*), (*b*), (*c*) and (*d*), show the patterns obtained in a vertical field. For (*a*) the disc was demagnetized by reversal of a slowly decreasing vertical field. The arrowhead deposits are of unequal intensity, alternate deposits being heavier than the others. On applying a field normal to the disc the lighter deposits became less dense and eventually disappeared completely on increasing the field. Figure 12(*b*) shows the effect at a field of 120 oersteds directed inwards, and 12(*c*) the change on reversal of this field. Evidently the disc was not completely demagnetized when 12(*a*) was obtained. The deposits shown in (*c*) became heavier and wider as the vertical outward field was still further increased; 12(*d*) shows the effects at 800 oersteds, where the narrow white regions are the positions of the deposits in 12(*b*).

Similar patterns were observed when a small field was applied parallel to the surface. Complete demagnetization in a horizontal field gave a pattern similar to Figure 12(*a*) but the deposits were of equal intensity and more widely spaced. It was also noted that the spacing varied considerably over the surface of the disc. Application of a small field nearly parallel to the lines caused the deposits to become alternately more and less dense, as shown in Figure 14, similar to the effect in a vertical field. A corresponding effect was also observed on reversing the horizontal field. However, none of the deposits disappeared entirely, but at about 10 oe. the pattern began to change completely. The arrowhead type deposits rapidly broke up and a pattern of straight lines at about 45° to them began to form. These were short at first, but, as the field increased they joined up to form longer lines, some of them extending right across the disc. They only formed at an angle greater than 45° with the field. On rotating the disc an alternative set of lines appeared when the angle became less than 45°; these lines were approximately at right angles to the original lines. The spacing of both sets of lines was of the order of $30\,\mu$, a distance which did not alter with the orientation of the disc, or show marked variation with field intensity. All these patterns can be interpreted on the views set forth by Bozorth, Williams and Shockley (1949), whose paper, together with those of Stoner (1950) and Kittel (1949) may be consulted for further information.

As this communication is mainly concerned with quantitative results, it is sufficient to state here that the 'arrowhead' deposits in Figures 12 and 14 are formed on those portions of the surface where the underlying domain magnetizations run vertically and alternately in opposite directions in the figures. There are small components of these magnetizations perpendicular to the surface, and these give rise to deposits which are either intensified or weakened on applying fields as in Figures 12 (*b*) and (*c*), and 14. The regions between the deposits are magnetized parallel to the surface and perpendicular to the lines. On applying a very strong vertical field the regions must decrease in extent, as in 12(*d*), where most of the disc is magnetized parallel to the normal field, but domains magnetized anti-parallel thereto are still present, as shown by the colloid-free areas which are of smaller extent. From the nature of the patterns one may deduce that one quaternary axis, say the [001], lies close to the surface of the disc but that neither the [010] nor the [100] axis is parallel to the surface. No apparent variation in spacing of the deposits with change in the horizontal field was observed, but it was seen that the spacing was somewhat less on regions remote from, than on those near to, the centre of the disc.

Disc No. 2. With disc No. 2, as with disc No. 1, it was not possible to determine the orientation accurately after etching with dilute nitric acid, though from measurements with the torque magnetometer, combined with observations on powder deposits, it was concluded that the [010] and [001] axes lay within 5° of the surface. The diameter of the disc was 1·2 cm. and its final thickness 0·18 mm. After lengthy electropolishing in orthophosphoric acid the surface was very uneven. At the edge of the disc in an applied (not effective) field of 3 oe., the pattern was similar in some respects to those obtained by Bozorth, Williams and Shockley (1949), and showed that this region of the surface was more nearly parallel to the (100) plane than in the case of the disc of Figures 12 and 14. Striations of heavy deposit parallel to the field indicated that the underlying domains were magnetized perpendicular to the field. However, calculation showed that the effective field could only be about 0·03 oersted on account of the large demagnetization factor for the disc, so that the pattern was practically characteristic of the demagnetized state. The closure domains magnetized at right angles to the sub-surface domains were clearly visible.

On rotating the disc in this field the pattern did not alter. Rotation of the disc by 45° from its last position in an anti-clockwise direction and increasing the effective field to about 1 oe. caused the 90° boundaries perpendicular to the field to lengthen and those parallel to the field to shorten. For about 10 oe., the transition was complete. The domain boundaries were now all of the 90° type and were all perpendicular to the field. The alternative set of 90° boundaries could be observed by rotating the disc through a further 90°, but they were not as clear as the previous set. The spacing of the lines decreased as the field increased, giving approximately the expected variation in *d*, but it was impossible to measure *d* accurately over a chosen small region. The spacing decreased in going from the centre to the edge of the disc but not as much as one would expect.

Disc No. 3. With disc No. 3 the crystal orientation was determined more accurately by developing etch pits with warm ferric sulphate solution. In order to produce a more even surface, the disc, 1 cm. in diameter, was given a preliminary hand polishing and a shorter electrolytic polishing in orthophosphoric acid. However, the spacing of line deposits perpendicular to a field applied along a binary axis was still very uneven, as with disc No. 2. Disc No. 3 was therefore used in a preliminary test to find which of the two types of boundary, (011) or $(01\bar{1})$, gave the most regular deposits. The strip used in the experiments described above was then cut with its long edge, chosen to be the direction also of the field, perpendicular to the direction of the best deposits.

From the results of the etch pit reflections and magnetometer measurements the angle between the [100] axis and the normal to the disc surface was found to be $4\frac{1}{2}$°. The [010] and [001] axes were inclined to the surface at about $6\frac{1}{2}$° and 6° respectively. The most regular deposits were obtained when the [011] axis, rather than the $[01\bar{1}]$ axis, was parallel to the field, and accordingly the strip was cut with its long edge parallel to the projection of the $[01\bar{1}]$ axis in the surface.

§ 8. DISCUSSION

The foregoing experiments have shown that within the limits of experimental error the average width of the primary domains in a single crystal strip of silicon-iron, magnetized parallel to a binary axis lying in the surface, decreases continuously as the magnetic field increases. The decrease is at first very rapid and finally becomes very slow in strong fields. The changes are approximately in agreement with Néel's calculations, but the average domain width does not exhibit a minimum value, and the discrepancies in the case of narrow strips are rather large. We have not sufficient evidence to allow us to say that the average domain width varies discontinuously with the field, for it is difficult to follow individual boundary changes with the powder technique as the colloid particles tend to stick to the surface. However, we know that in the fundamental experiment on a single 180° boundary carried out by Williams and Shockley (1949) the displacement was surprisingly regular and in accord with theory.

With improvements in apparatus and better choice of specimens it should be possible to get more accurate information on domain structures and wall movements, and their dependence on specimen shape, crystal orientation, field strength and mechanical strain. It would be advantageous to obtain thicker specimens, so that the surface might be ground accurately parallel to a chosen plane without reducing the thickness of the material too much, and to take more precautions to ensure that the field acting within a specimen is uniform.

ACKNOWLEDGMENTS

The silicon–iron specimens used in this work were very kindly supplied by Mr. G. Richer, Orb Iron Works, Newport, Mon. We are indebted to the British Electrical and Allied Industries Research Association for financial support which made this work possible, and for permission to publish the results.

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The Infra-red Spectrum of Magnesium Oxide *

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MS. received 27th July 1949, *and in amended form* 21*st September* 1949

ABSTRACT. An investigation of the infra-red spectrum of magnesium oxide has been carried out in the range up to 25·5 μ, both in reflection and transmission. The eigenfrequency is found to be at 17·5 ± 0·3 μ and four subsidiary maxima have been found in transmission. The results of Barnes, Brattain and Seitz in the range up to 15·6 μ have not been confirmed and no detailed fine structure has been found. The reflection spectrum of magnesium oxide has been fully investigated up to 25·5 μ, and a broad reflection maximum has been found extending from 17 μ to 21 μ, with subsidiary maxima at 14·02 μ and 23·8 μ.

From the data obtained curves have been drawn to show the behaviour of the two optical constants n and k in the range from 7 μ to 25 μ.

§ 1. INTRODUCTION

The early theoretical work carried out by Born and others on the behaviour of polar crystals in the infra-red predicted that crystals with cubic symmetry should show only one absorption maximum, together with a corresponding reflection maximum in the same frequency region. The first complete and reliable test of this theory was provided by the results of Czerny (1930) and Barnes and Czerny (1931) on the absorption of sodium chloride and potassium chloride in the infra-red: the main work was carried out on sodium chloride. Contrary to expectation it was found that, apart from the main absorption maximum at 61·1 μ there existed subsidiary maxima at 40·5 μ and 51·0 μ. Likewise, besides the main reflection maximum at 52 μ, subsidiary maxima were found at 38 μ and 48 μ. Similar results were obtained for potassium chloride.

Barnes (1932) extended the work to some fifteen alkali-halide crystals, and in every case a subsidiary maximum was found on the short wavelength side of the main absorption maximum.

Born and Blackman (1933) attempted an explanation of these results in terms of anharmonic forces between particles. They showed that, for a linear chain of particles, two subsidiary maxima were to be expected, one on the short wavelength side and one on the long wavelength side of the main maximum. The former agreed reasonably well in position with the subsidiary maximum found at 40·5 μ in the case of sodium chloride. Later Blackman (1933, 1936) extended the work to the three dimensional lattice and showed that subsidiary maxima could also occur elsewhere.

An inherent difficulty in all the work on these alkali-halides was the fact that their eigenfrequencies were well out in the infra-red, where the amount of radiation available is very small. Therefore, in order to obtain a reasonable deflection of the recording instrument, the spectrometer must have very wide slits and hence low resolving power. Consequently attention was turned to substances of the same crystal structure but with their main absorption at much shorter wavelengths. Of these magnesium oxide is the most suitable, largely because of its availability in large crystals.

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* This paper forms part of a thesis for the degree of Ph.D. at London University.

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The earliest relatively complete study of magnesium oxide was made by Tolksdorf (1928), who examined its transmission properties in the range from 2 μ to 15 μ, with finely powdered magnesium oxide held between plates of rock-salt. The transmission for these specimens varied from 40% at 2 μ to 95% at 15 μ, with minima of transmission at 3·85 μ, 7·65 μ and 14·2 μ. The low value of the transmission at 2 μ indicates that the particle size was much too great. This would lead to spurious results and, indeed, no later worker has confirmed these results.

Strong (1931) investigated the transmission of layers of magnesium oxide formed by burning magnesium ribbon, using reststrahlen. The results suggested a strong absorption around 22·9 μ.

Further work on magnesium oxide was carried out by Fock (1934) and Barnes, Brattain and Seitz (1935). The latter authors were the first to study magnesium oxide in the single crystal state. They obtained from the Norton Company artificially made single crystals from which they were able to cleave plates as thin as 75 μ. With these plates, and using a rock-salt prism spectrometer, they studied magnesium oxide both in transmission and reflection as far as 15·6 μ. In transmission they found some forty fine absorption lines in the range from 6 μ to 15·6 μ. The number and position of these lines varied with the thickness of the specimen. Such a fine structure was hitherto unknown for face-centred cubic crystals, though it had been reported for quartz and calcite (Silverman and Hardy 1931, Silverman 1932). They also compared these results with those obtained with layers formed by burning magnesium ribbon, and by evaporating magnesium oxide powder in a high vacuum. While the results obtained agreed in a general way with the results obtained using thin cleaved plates, inasmuch as both sets showed considerable fine structure, there appears to be no detailed correspondence, line for line, between the two sets of results, though Barnes *et al.* claim agreement in almost every detail.

The results obtained for the reflection power of magnesium oxide are also somewhat unusual. A double reflection maximum was found, the peaks being at 14·8 μ and 15·4 μ. They were both sharp, and gave values of reflection of 63% and 66% respectively. At 15·6 μ the reflecting power had dropped to 20%.

In order to explain their results the authors advanced a quantum mechanical theory based on an anharmonic potential function and the interaction of crystal vibrations as originally suggested by Born and Blackman. However, the assumptions which lead to the detailed fine structure do not appear to be correct (Blackman 1936).

The results of Barnes, Brattain and Seitz were in complete contradiction to those of Fock (1934), of whose work they did not appear to be aware. Fock studied the transmission of fumed layers of magnesium oxide up to 40 μ. He reported the eigenfrequency to be at 17·3 μ, with subsidiary maxima at 14·9 μ, 6·8 μ and 25 μ, but no fine structure. He states that his specimens were of the order of 1 μ in thickness, and the evaporated specimen of this thickness used by Barnes, Brattain and Seitz shows as much fine structure as any of their specimens. Further, Fock shows all his points on his curves and gives slit widths, whereas Barnes, Brattain and Seitz show neither.

In view of this contradiction of the results obtained by different workers, and the theoretical work of Barnes, Brattain and Seitz, a fresh investigation with adequate resolving power appeared desirable both from a theoretical and from a practical standpoint.

§ 2. EXPERIMENTAL

A prism spectrometer was used for the experimental study of the infra-red spectrum of magnesium oxide. Two prisms were available, one of natural rock-salt for use up to 15 μ, and one of artificially grown potassium bromide for use up to 26 μ. Both prisms had faces approximately 10 cm. square, and had a refracting angle of 60°.

Artificially produced single crystals of magnesium oxide were again obtained from the Norton Company. One of these crystals had a good cleaved surface on one side some 5 cm. long and 1·8 cm. wide, and this was used for the reflection measurements.

The radiation was detected by means of a thermopile and the resultant E.M.F. was measured by means of a galvanometer and photoelectric amplifier system.

The layout of the spectrometer for the transmission measurements is shown in Figure 1; the specimen, mounted in a sliding tower, is introduced into the path of the radiation directly in front of the thermopile. The layout for the reflection measurements is shown in Figure 2 up to the entrance slit S, and beyond this in Figure 1. The sliding tower R is introduced at the point where the mirror M 4 brings the radiation to a focus, so that an image of the source is formed either on a plane front silvered mirror or on the plane surface of a crystal of magnesium oxide. The angle of incidence was approximately 10°.

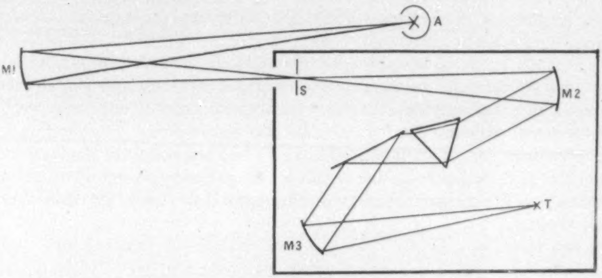

Figure 1.

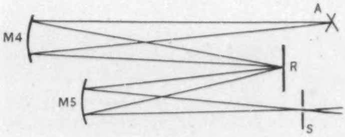

Figure 2.

The Source.

The source of radiation A (Figures 1 and 2) was an upright Welsbach mantle surrounded by a brass cylinder with a slit down the middle through which the radiation emerged. A gas mantle was used in preference to a Nernst filament as it gives more radiation at the longer wavelengths and, since it does not have the very high intensity peak at 1·5 μ shown by a Nernst filament, there is less danger of contamination of the longer wavelengths due to scattering of the shorter wavelength radiation. Preliminary experiments showed there was no significant inaccuracy due to scattering when a gas mantle was used, and also gave confirmation of the wavelength calibration since the same percentage transmission was obtained in the region of 14μ, irrespective of the prism used.

Production of Specimens.

Plates of magnesium oxide were cleaved from a massive crystal, and in this way specimens down to 76μ thickness could be produced. The thickness of these plates was found by measuring their area and weighing them.

The thinnest of the plates produced in this way was completely opaque from $13{\cdot}5\mu$ to the limit of the spectrometer (see Figure 3), and another method of producing yet thinner layers had therefore to be used. Since magnesium oxide is too hard to be shaved down in the way used by Czerny (1930) for producing thin deposits of rock-salt, it was decided to produce thin deposits of magnesium oxide by burning magnesium ribbon and collecting the vapour on collodion films.

The layers produced in this way consist of an agglomeration of minute crystals which will appear quite homogeneous to radiation of wavelength 10μ and longer. It was not possible to measure the thickness of these layers directly or by weighing, and the actual determination by calculation is discussed in a later section.

To confirm the results found with the fumed layers and to demonstrate the coherence of these layers in this region an evaporated specimen was prepared. Kirchner (1932) has shown by electron diffraction that such evaporated layers are single crystals.

§ 3. METHOD OF CALCULATION

In order to calculate the refractive index n and the extinction coefficient k of a medium from experimental data at any given wavelength, it is necessary to know the reflection coefficient R for a large crystal, and the transmission coefficient D for a specimen of known thickness. In practice the formulae are too complicated to allow explicit expressions for n and k to be obtained, and one has to resort to methods of successive approximation. When the wavelength is reasonably removed from the position of main absorption, however, the reflection coefficient is chiefly dependent on the refractive index, and the transmission coefficient depends mainly on the extinction coefficient. The process of approximation is then fairly rapid.

The Reflection Coefficient.

The reflection R from a single plane surface of a substance, with optical constants n and k, is given, at normal incidence, by the formula

$$R=\frac{(n-1)^2+k^2}{(n+1)^2+k^2}. \qquad \ldots\ldots(1)$$

Solving this for n, we obtain

$$n=\frac{1+R}{1-R}\pm\left[\frac{4R}{(1-R)^2}-k^2\right]^{1/2}. \qquad \ldots\ldots(2)$$

As n is always real it follows that k^2 is always less than $4R/(1-R)^2$ and so one may expand the expression in ascending powers of k^2, obtaining

$$n_1 = \frac{1+\sqrt{R}}{1-\sqrt{R}} - \frac{k^2}{4}\left(\frac{1-R}{\sqrt{R}}\right) - \frac{k^4}{64}\left(\frac{1-R}{\sqrt{R}}\right)^3 - \dots \qquad \dots\dots(3)$$

$$n_2 = \frac{1-\sqrt{R}}{1+\sqrt{R}} + \frac{k^2}{4}\left(\frac{1-R}{\sqrt{R}}\right) + \frac{k^4}{64}\left(\frac{1-R}{\sqrt{R}}\right)^3 + \dots \qquad \dots\dots(4)$$

As k is usually considerably less than one (and where it is greater than one R is very large) these expressions converge very rapidly. In only a very few cases was the third term significantly different from zero.

Both the values of n given can have a physical significance and the correct value cannot be decided from the reflection power alone, but it is known in general that the refractive index is greater than unity on the short wavelength side of the reflection minimum, and less than unity between the reflection minimum and the main absorption maximum.

The Transmission Coefficient.

The transmission D of a plane parallel plate is given (Kellner 1929, Czerny 1930) by

$$D = \exp(-4\pi kd/\lambda)\frac{(1-R)^2 + R\sin^2\psi}{[1-R\exp(-4\pi kd/\lambda)]^2 + 4R\exp(-4\pi kd/\lambda)\sin^2(\alpha+\psi)} \qquad \dots\dots(5a)$$

where $\alpha = 2\pi nd/\lambda$ and $\tan\psi = 2k/(n^2+k^2-1)$.

To calculate k from R and D it is more convenient to rearrange (5 *a*) as follows:

$$[\exp(2\pi kd/\lambda) - R\exp(-2\pi kd/\lambda)]^2 = \frac{(1-R)^2 + 4R\sin^2\psi}{D} - 4R\sin^2(\alpha-\psi). \qquad \dots\dots(5b)$$

It is often possible to neglect the term $R^2\exp(-4\pi kd/\lambda)$ on the left hand side of (5 *b*) and we then have

$$\exp(4\pi kd/\lambda) = \frac{(1-R)^2 + 4R\sin^2\psi}{D} + 2R - 4R\sin^2(\alpha+\psi). \qquad \dots\dots(6)$$

Also it is frequently possible to neglect the term $4R\sin^2\psi$, and writing for the interference term its mean value $2R$ we get

$$\exp(4\pi kd/\lambda) = (1-R)^2/D. \qquad \dots\dots(7)$$

Which of the above formulae will be sufficiently accurate in any particular case depends on the magnitudes involved, and each case must be considered individually.

For the thicker plates, where the specimen thickness is many times the wavelength of the radiation, the value of α changes very rapidly. A detailed investigation showed that for specimens A, B and C the change in α over the waveband admitted by the slit width was sufficient to smooth out the interference effects, and thus equation (7) could always be used. This was not quite the case for specimen D, and, in view of the uncertainty in α due to its thickness, it was considered inadvisable to use the results obtained with this specimen in the region of high transmission for the calculation of k. Beyond $10\,\mu$ the increase in absorption and the fall in reflecting power make the interference term negligible

Calculation with the Fumed Specimens.

As there was no way of measuring the thicknesses of the fumed specimens these had to be calculated from the known values of n and k. The calculation of the thickness of magnesium oxide for specimen E (Figure 3) is simple, since at 12·15 μ the reflection coefficient R is less than 1% and n is very nearly unity, so that formula (7) may be used to calculate k from specimen D. With this value for k, the thickness of specimen E can then be calculated from its observed transmission. From the value obtained one can then calculate n and k for other wavelengths and thus obtain the thickness of the other specimens.

Calculations carried out at 11·7 μ and 12·15 μ gave nearly the same thickness for specimen E, viz. $d = 7{\cdot}75\,\mu$ and 7·85 μ (the agreement is closer than can be justified by the experimental data).

When the longer wavelengths are reached, where the reflection coefficient becomes appreciable, the fact that these fumed specimens contain a considerable amount of air has to be taken into account. This changes the effective constants n and k. The calculations at 11·7 μ and 12·15 μ give the thickness of magnesium oxide, i.e. the thickness the layer would have it if were compressed so as to exclude all the air. The amount of air present was estimated by calculating the value of k at three different wavelengths, using the results for specimen D. Then the thickness for specimen E was calculated for different amounts of air in each case. In these calculations the following assumptions were made:

(i) The effective refractive index n' is equal to the optical path length divided by the physical path length. Hence if the layer consists of, say, x parts by volume of magnesium oxide and one part of air, n' is given by

$$n' = (xn + 1)/(x + 1),$$

where the refractive index of air is taken as unity.

(ii) The effective value of the extinction coefficient k' for this layer is given by

$$k' = xk/(x + 1).$$

(iii) The effective reflection coefficient for this layer is given by

$$R' = \frac{(n' - 1)^2 + k'^2}{(n' + 1)^2 + k'^2}.$$

The results obtained for various amounts of air are shown in Table 1.

Table 1. Thickness (in μ) of Specimens E calculated for various Proportions of Air

Proportion of air	None	10%	20%	30%	40%
Wavelength :					
12·75 μ	—	—	8·44	7·76	6·98
13·0 μ	—	—	7·51	7·70	6·99
13·25 μ	(−2·21)	2·46	5·67	7·56	8·03

The amount of air is given as a percentage by volume of the calculated thickness of magnesium oxide.

It will be seen from the above results that if it is assumed that there is a quantity of air present equal to 30% of the magnesium oxide, results for d are obtained which are consistent both with each other and with the values found at $11{\cdot}7\,\mu$ and $12{\cdot}15\,\mu$, where the reflection is very small.

A further justification of this method of procedure is provided by the fact that at $17{\cdot}5\,\mu$ the mean value of k, calculated from the fumed specimens, is 2·07, and from the evaporated specimen $k = 2{\cdot}3$. In view of the uncertainty in the measurement of the thickness of the evaporated specimen this is a very satisfactory agreement.

§ 4. RESULTS

The results obtained for the infra-red spectrum of magnesium oxide are shown in Figures 3 and 4. Figure 3 consists of curves of percentage transmission for various thicknesses of specimen, and Figure 4 gives the reflection coefficient for a large crystal as a percentage of the corresponding reflection from a front-silvered plane mirror. The centres of the vertical lines on the curves represent the actual values obtained, and the lengths of these lines indicate the experimental error.

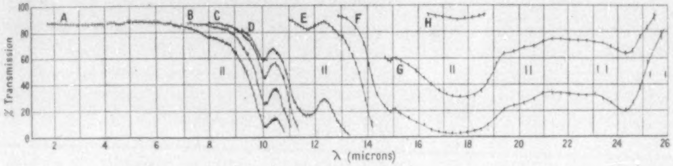

Figure 3.

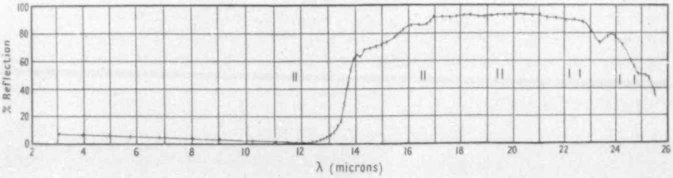

Figure 4.

Thickness of specimens

Specimen A 0·468 mm.	Cleaved from a massive crystal.	Specimen E $7{\cdot}80 \times 10^{-3}$ mm.	Fumed.
Specimen B 0·235 mm.		Specimen F $2{\cdot}67 \times 10^{-3}$ mm.	
Specimen C 0·124 mm.		Specimen G $1{\cdot}23 \times 10^{-3}$ mm.	
Specimen D 0·076 mm.		Specimen H $0{\cdot}19 \times 10^{-3}$ mm.	Evaporated.

The vertical lines on the curves indicate the observed points, together with the experimental error. The pairs of vertical lines indicate the width of the wavelength band received by the thermopile.

Each of the points shown on the curves of Figures 3 and 4 is the mean of two results obtained from separate runs through the spectrum, except in the case of the transmission curves beyond $23\,\mu$ where each point is the mean of five readings taken consecutively. In no case did the results obtained from the separate runs differ from each other by more than 2%. Furthermore, the result for each run is the mean of three readings taken consecutively; the standard deviation for these three results was in almost every case less than 1%.

The first portion of the work to be carried out was a very careful study of the percentage transmission in the region up to $14{\cdot}5\,\mu$. Readings were taken at a mean separation of 0·08 μ. This was done because of the startling results found in this region by Barnes, Brattain and Seitz (1935) referred to above. In view of the contradiction between the results found by these authors and those presented here, it seems desirable, before proceeding to a more general consideration of the results, to give a detailed comparison of the two sets of results.

Comparison with the Results of Barnes, Brattain and Seitz.

The chief feature of the transmission results presented by Barnes, Brattain and Seitz was the number of fine absorption lines in the range from 6 μ to 15·6 μ. These lines varied in depth between about 1% change in absorption, and 10 to 15% change. The numbers also varied considerably with the thickness of the specimen, as did their position and intensity. A test of the consistency of the results obtained is given by the values of k calculated from different thicknesses of specimen. In Tables 2 and 3 are given values of k for different thicknesses of specimen, calculated from the results obtained here, and from the results obtained by Barnes, Brattain and Seitz. In each case, in the final column, the quantity $(k_{\max}-k_{\min})/k_{\text{mean}}$ is given as a percentage for each set of values obtained at each wavelength. Both sets of results were obtained using specimens cleaved from artificially grown crystals of magnesium oxide obtained from the Norton Company.

Table 2. Values of $k \times 10^3$ calculated from the Results of the Present Work

	$k \times 10^3$				$k_{\text{mean}} \times 10^3$	$\frac{k_{\max}-k_{\min}}{k_{\text{mean}}}$ (%)
d (mm.)	0·468	0·235	0·124	0·076		
λ (μ)						
9·00	0·57	0·56	0·62	—	0·58	10
9·25	0·81	0·90	0·91	—	0·87	11
9·50	1·25	1·35	1·39	—	1·33	10
9·75	2·17	2·22	2·27	—	2·22	5
10·00	4·07	4·15	4·56	—	4·26	11
10·25	3·70	3·86	3·94	4·01	3·88	8
10·50	3·31	3·35	3·48	3·93	3·52	18
10·75	5·07	5·23	4·96	5·54	5·20	11
11·00	—	7·83	8·57	8·90	8·43	13
11·25	—	—	15·8	15·6	15·7	2

Table 3. Values of $k \times 10^3$ calculated from the Results of Barnes, Brattain and Seitz

	$k \times 10^3$				$k_{\text{mean}} \times 10^3$	$\frac{k_{\max}-k_{\min}}{k_{\text{mean}}}$ (%)
d (mm.)	1·75	0·47	0·12	0·075		
λ (μ)						
9·0	0·39	0·72	0·63	—	0·58	41
9·5	0·70	1·04	0·98	—	0·91	37
10·0	—	3·13	4·12	2·62	3·29	46
10·5	—	—	5·75	3·01	4·38	62
11·0	—	—	4·90	4·18	4·54	16
11·5	—	—	∞	12·3	—	—
12·5	—	—	16·7	11·2	13·9	40

It will be seen that the results given by Barnes, Brattain and Seitz are not nearly so self-consistent as those presented here, but that the mean values of k are of the same order of magnitude up to 10·5 μ. From this point onwards the discrepancy becomes very much more marked. As can be seen from their curves, these authors found that a specimen of 0·075 mm. thickness transmitted between 10 % and 30 % of the radiation right up to 15·6 μ, while a similar specimen of 0·076 mm. was found here to be opaque from 13·5 μ to beyond 25 μ. To obtain 35 % transmission at 15·6 μ it was found necessary to go down to a thickness of 1 μ to 2 μ. This suggests that there may have been considerable scattering of the radiation in their apparatus.

Further, comparing the results obtained in reflection, Barnes, Brattain and Seitz found the minimum reflection to be about 5 %, compared with a value of less than 0·5 % found here, at 12·15 μ. In a paper on Christiansen filters, Barnes and Bonner (1936) reported that a deposit of fairly coarsely powdered magnesium oxide on a very thin film gave a maximum transmission of more than 60 % at 12·2 μ. For this to occur the reflection coefficient at this point must be negligible. In the same paper Barnes and Bonner give the refractive index of magnesium oxide at 9·0 μ as 1·43, which gives a reflection coefficient of 3·1 %, compared with 3·0 % found here, and 5·5 % found by Barnes, Brattain and Seitz. Also the double peak in reflection of 63 % at 14·8 μ and 66 % at 15·4 μ found by Barnes, Brattain and Seitz is not confirmed by the present work.

In view of the above considerations, and the fact that the results shown here are a direct confirmation of the work of Fock, it can be asserted that the infra-red spectrum of magnesium oxide does not possess a fine structure of the nature reported by Barnes, Brattain and Seitz.

It should be mentioned that the results of Barnes, Brattain and Seitz have been used by Raman (1947) in support of his theory of lattice vibrations. The various lines they found are attributed to combinations between the various 'fundamental frequencies of the lattice'. As their results are not confirmed the infra-red spectrum of magnesium oxide can no longer be considered as supporting Raman's theory.

General Discussion of the Results.

The main absorption maximum for magnesium oxide was found to be at 17·5 ± 0·3 μ, in good agreement with the value of 17·3 ± 0·3 μ found by Fock (1934). Subsidiary maxima are seen to occur at 10·07 μ, 11·7 μ and 14·8 μ on the short wavelength side of the main absorption, and at 24·3 μ on the long wavelength side. Two of these subsidiary maxima have previously been reported by Fock, except that he found the subsidiary maximum on the long wavelength side to be at 25 μ, but the two maxima at 10·07 μ and 11·7 μ are reported here for the first time. Fock did not use sufficiently thick specimens to observe these two. The subsidiary maximum found by Fock at 6·8 μ was not observed here and is considered to be spurious, and probably due to absorbed moisture. This view is supported by the fact that a specimen of crystalline magnesium oxide, left out in the open for some weeks, developed a cloudy surface and showed a sharp maximum of absorption at 6·4 μ. This vanished after the specimen was cleaned by rubbing it gently with a piece of clean dry chamois leather until the surfaces were quite clear again.

The main reflection maximum was found to be very broad and to extend from 17 μ to 21 μ, the value of R being between 92 % and 93·5 % over the whole of this range. Subsidiary maxima were found at 14·02 μ and 23·8 μ, and a flattening of the curve was observed at 16·4 μ and 25 μ. It is clear that the two subsidiary maxima at 14·02 μ and 23·8 μ are associated with the absorption maxima that occur at 14·8 μ and 24·3 μ. As is commonly found in these cases, with the exception of the main maximum, the reflection maxima lie on the short wavelength of the corresponding absorption maxima. The small flattening in the reflection curve between 16·1 μ and 16·7 μ is somewhat unusual, but has been carefully checked. There appears to be no corresponding variation in the transmission curve. The flattening in reflection at 25 μ suggests there may be anomalies in the absorption spectrum beyond the range of the spectrometer used in the present case.

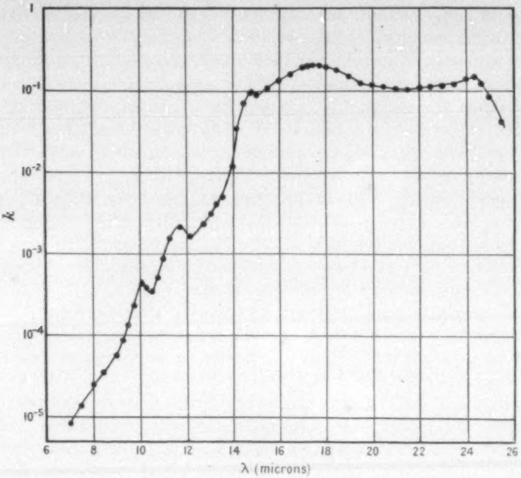

Figure 5.

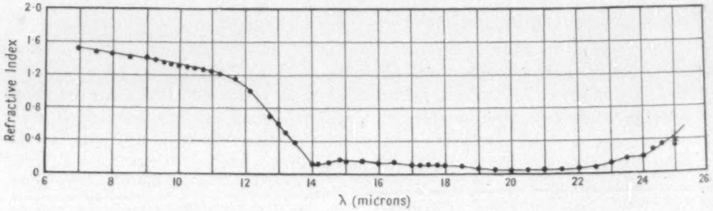

Figure 6.

The refractive index and extinction coefficient for magnesium oxide have been calculated from the curves for transmission and reflection, and the results of these calculations in the region fron 7 μ to 25·5 μ are shown in Figures 5 and 6. The curve for the extinction coefficient shows very clearly the positions and relative intensities of the various absorption maxima. The very rapid fall of k on the short wavelength side of the eigenfrequency should be noted, and compared with the similar results found by Czerny (1930) on rock-salt. The main point to be noted in the refractive index curve is the fact that the refractive index remains very low right out to 25 μ. With most crystals of the type considered here the reflection coefficient has fallen to about 80% or less at the main absorption maximum, and the refractive index rises rapidly, passing through the value unity near the eigenfrequency, and reaching values of 6 to 10 on the long wavelength side. After reaching a peak the refractive index then falls towards the value given by $\sqrt{K}$, where K is the static dielectric constant. However, a consideration of equations (2), (4) and (5) shows that, of the two values for n given by equation (2), only the lower one is possible in the whole of the region beyond the eigenfrequency investigated here. The beginning of a rise is clearly shown in Figure 6, but the value at the last point (circle) has decreased again. The application of a slit width correction to the results obtained in this region had the effect of lowering slightly the transmission curves in the region of 25 μ, but did not change the reflection curve. This was just sufficient to raise the refractive index to a value of 0·37, indicated by a cross in Figure 5.

That the refractive index does eventually rise to a value of $\sqrt{K}$ is shown by the results of Strong (1931), who studied the reflection of magnesium oxide in the range from 20·7 μ to 152 μ, using reststrahlen. The first two values at 20·7 μ and 23 μ are somewhat uncertain since the line indicating 80% reflection on Strong's graph appears to be misplaced. However it seems that the values he obtained are about 10% below the values found here, probably because he used reststrahlen. There are values of 44% at 27·3 μ and 42% at 29·4 μ, and then a steady decrease in reflecting power from about 35% at 32·8 μ to 29% at 152 μ. If we use Højendahl's (1938) value of 9·8 for the dielectric constant of magnesium oxide then, with $n = \sqrt{K}$, we find $R = 27\%$.

If we assume, from Strong's results, that in the region of 33 μ the refractive index has a value of about four, then somewhere between 25 μ and 33 μ the refractive index will pass through the value unity. As the extinction coefficient has already fallen below 0·5, this means that the reflection coefficient must fall to a very low value (less than 5%) at this point. One would expect such a pronounced anomaly in the reflection curve to have its counterpart in the absorption spectrum. A careful study of Fock's results in this region suggests that there may be a further small subsidiary maximum at about 32·5 μ. Clearly a further investigation of this region, both in transmission and reflection, would be very desirable.

Comparison with other Face-centred Cubic Crystals.

All other workers investigating the absorption of face-centred cubic crystals have found subsidiary maxima on the short wavelength side of the eigenfrequency, but only in the case of lithium fluoride (Barnes 1932) has a subsidiary maximum been found on the long wavelength side and fairly close to the main absorption. Even so, it is extremely doubtful if this is a genuine subsidiary maximum as it falls exactly in the position of the main maximum of absorption of sodium fluoride and is probably due to a small quantity of sodium fluoride contained in the lithium fluoride.

On the short wavelength side it is found that if the wavelength of the main maximum is, in turn, divided by each of the wavelengths of the subsidiary maxima on the short wavelength side, then for rock-salt, potassium chloride and magnesium oxide the results shown in Table 4 are obtained. From these results it would appear that there are considerable similarities in the vibrational spectra of these three substances. In most of the cases investigated by Barnes (1932) only one subsidiary maximum was found, the ratio being about 1 : 1, perhaps in many cases because the shorter wavelengths were not investigated.

Table 4

Substance	λ main/λ subsidiary
NaCl	1·18, 1·50, 1·80
KCl	1·18, 1·50, 1·67, 1·73, 2·14
MgO	1·18, 1·50, 1·74

Parodi (1937, 1938) has carried out an investigation of other oxides, using reststrahlen. Of the face-centred cubic crystals investigated cobalt oxide and cadmium oxide both had a subsidiary maximum on the short wavelength side of the eigenfrequency. The ratio of the wavelengths of the two maxima was 1·38 for cobalt oxide and 1·29 for cadmium oxide. No subsidiary maxima were observed on the long wavelength side of the eigenfrequency in either case.

The reflection spectrum for magnesium oxide shows a marked dissimilarity from the reflection spectra of such salts as sodium chloride, potassium chloride, lithium fluoride. In the first place there is no well-defined maximum. Secondly the centre of this range lies on the long wavelength side of the eigenfrequency, while in the cases mentioned the peak of reflection occurs on the short wavelength side of the eigenfrequency. Finally there is the subsidiary maximum in reflection at 23·8 μ.

All these differences are probably closely bound up with the existence of the very strong subsidiary maximum which occurs in absorption at 24·3 μ, and the fact that it is so close to the main absorption.

Thus while, in general, magnesium oxide shows many features similar to those found for other face-centred cubic crystals, it is differentiated from them in absorption by its behaviour on the long wavelength side of the eigenfrequency, and in reflection by the extent and position of its region of high reflecting power.

§ 5. COMPARISON OF RESULTS WITH THEORY

The Eigenfrequency.

The eigenfrequency of a cubic crystal is related to the other properties of the crystal by the equation (Born and Goeppert-Mayer 1932)

$$\frac{K-K_0}{[1+\tfrac{1}{3}\gamma(K_0-1)]^2}=\frac{Ne^2}{\pi M\omega^2 a^3}, \qquad \ldots\ldots(8)$$

where K is the static dielectric constant, K_0 is that portion of K due to the polarizability of the ions, and may be put equal to n^2 where n is the optical refractive index, e is the ionic charge, N is the number of atoms per cell, M is the reduced mass, a is the distance between next neighbours in the lattice, ω is the eigenfrequency and γ is a constant depending on the overlap between adjacent ions. The denominator on the left hand side is the term introduced by consideration of the Van der Waals forces produced by the polarizability of the ions; $\gamma=1$ corresponds to no overlap between adjacent ions, so that the field polarizing any ion is $E+4\pi P/3$. Mott and Littleton (1938) give reasons for supposing that γ is in fact nearer zero.

Unfortunately the values given in the literature for the dielectric constant of magnesium oxide vary between rather wide limits. Günterschulze and Keller (1932), using the method of mixtures, give a value of 8·2, and Højendahl (1938), using an immersion method obtained values between 9·25 and 9·93, with a value of 9·80 considered as the most probable. Furthermore, the above formula is deduced on the assumption of central forces between the particles, and for central forces the elastic constants c_{12} and c_{44} should be equal. In magnesium oxide the ratio is about two to one (Durand 1936). If formula (8) is used to determine γ we get values of $\gamma=0{\cdot}64$ using Højendahl's value for K, and $\lambda=0{\cdot}37$ using the value given by Günterschulze and Keller. These are similar to values obtained for other crystals.

The Subsidiary Maxima.

According to the theory of Born and Blackman (Born and Blackman 1933, Blackman 1933, 1936) subsidiary maxima are the result of the coupling of the eigenfrequency to two other vibrations of the crystal lattice, both of which have the same wave number. Absorption will then take place at frequencies (called the combination frequencies) equal to the sum and difference of the two frequencies thus coupled to the eigenvibration. To obtain high absorption a large number of these combination frequencies must have the same value, and the observed subsidiary peaks represent the peaks in the spectrum of the combination frequencies.

Blackman (1936) has considered the case of a three-dimensional cubic lattice in which each atom is assumed to interact with its twenty-six nearest neighbours. Thus he does not consider the long range electrostatic forces. He assumes that the maximum density of vibrations occurs for the wave number where the frequency branches of the lattice vibrations split on passing from the case of equal masses to that of unequal masses. In this way Blackman obtained an expression for the frequencies of the subsidiary maxima which can be evaluated in terms of the eigenfrequency and the elastic constants of the substance. Comparison with the case of magnesium oxide shows fairly good agreement on the short wavelength side, the calculated absorption frequencies falling into those groups close to the observed subsidiary maxima. On the long wavelength side there is no agreement, but this might be expected as the difference terms are very much more sensitive to variations in the different branches of the crystal vibrations. To obtain better agreement it would be necessary to calculate the frequency spectrum of magnesium oxide. Kellermann (1940) has done this for rock-salt, but comparison with experiment is difficult as he does not order the various frequencies, for a given wave number, according to their type, but according to their magnitude. As far as can be judged the agreement is no better than that obtained by Blackman.

§ 6. CONCLUSION

Magnesium oxide has its eigenfrequency at $17{\cdot}3\,\mu$ and various subsidiary maxima, but does not show the fine structure as reported by Barnes, Brattain and Seitz. There is at least one subsidiary maximum on the long wavelength side of the eigenfrequency which is observed both in reflection and transmission. The reflection spectrum is unlike that of other face-centred cubic crystals, and has various anomalies on the long wavelength side of the eigenfrequency.

ACKNOWLEDGMENTS

The author wishes to thank Dr. L. Kellner for suggesting the problem and supervising the research, which was carried out in the spectroscopic department of the Imperial College directed by Dr. R. W. B. Pearse. He also wishes to thank Dr. M. Blackman for helpful discussions.

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LETTERS TO THE EDITOR

The Photo-disintegration of Carbon into Three Alpha-particles

Hänni *et al.* (1948) have recently studied the photo-disintegration of carbon into three alpha-particles, using lithium gamma-rays (~17·6 Mev.). The disintegration was found to proceed in two stages, via the well known 3 Mev. level in ^{8}Be (Hornyak and Lauritsen 1948), and may be described by the reactions

$$^{12}\mathrm{C} + h\nu \rightarrow {}^{8}\mathrm{Be}^{*} + {}^{4}\mathrm{He}, \qquad \ldots\ldots (1)$$

$$^{8}\mathrm{Be}^{*} \rightarrow {}^{4}\mathrm{He} + {}^{4}\mathrm{He}. \qquad \ldots\ldots (2)$$

In the investigation reported in this note, Ilford C2 and E1 nuclear emulsions were irradiated with a continuous spectrum of gamma-rays, of peak energy approximately 25 Mev., from the A.E.R.E. synchrotron, and the resulting 'carbon stars' were measured. For each star, the incident gamma-quantum energy was determined from the equation

$$h\nu = (E_{\mathrm{T}} + 7{\cdot}3)\ \mathrm{Mev.}, \qquad \ldots\ldots (3)$$

where E_{T} is the sum of the energies of the three alpha-particles. The stars were checked for correct momentum balance to avoid confusion with reactions such as ^{12}C(n, n)3 ^{4}He

(Green and Gibson 1949), or $^{16}O(\gamma, 4\alpha)$ (Goward, Titterton and Wilkins 1949). The latter reaction gave small four-particle stars, but since two of the tracks were often very close together there was considerable danger of mistaking them for three-particle stars.

Figure 1 shows a histogram, (a), of the total energy release, E_T, of 216 stars. From the known irradiation of the emulsions, monitored by a thick-walled ionization chamber, and a theoretical spectrum (Heitler 1944), the superposed cross section values, (b), may be deduced. The cross section has a maximum of the order of 10^{-28} cm² at about 18 Mev. gamma-ray energy and the effective threshold appears to be well above that calculated from the mass values, in agreement with reactions (1) and (2) and the potential barrier effect. The fall of the cross section above about 18 Mev. may be accounted for qualitatively either by (γ, p) and (γ, n) competition or in terms of an alpha-particle model as studied by Telegdi and Verde (1949). The apparent tendency for the cross section to rise above about 21 Mev. is not well established but is possibly correlated with the excitation of levels other than the 3 Mev. level in ^{8}Be (see below).

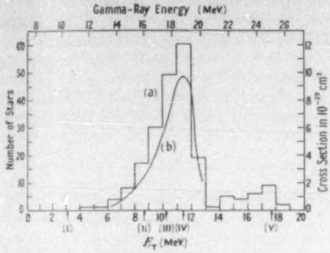


Figure 1.

- (*a*) Total energy released (E_T) in 216 carbon stars.
- (*b*) Approximate cross section, deduced from (*a*). Difficulties in the measurement of gamma-ray intensities, in addition to various other factors, introduce uncertainties of some ±50%.

Arrows on the E_T scale indicate:

- (i) The expected threshold, allowing for 3 Mev. ^{8}Be excitation.
- (ii) The $^{12}C(\gamma, p)^{11}B$ reaction threshold.
- (iii) The effective (γ, p) threshold allowing for potential barrier effect.
- (iv) The $^{12}C(\gamma, n)^{11}C$ reaction threshold.
- (v) The gamma-ray spectral limit (uncertain to ±0·5 Mev.).

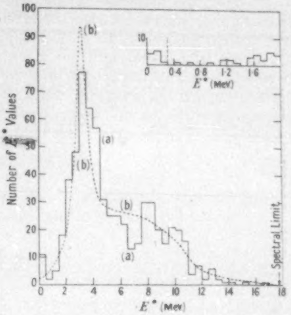


Figure 2.

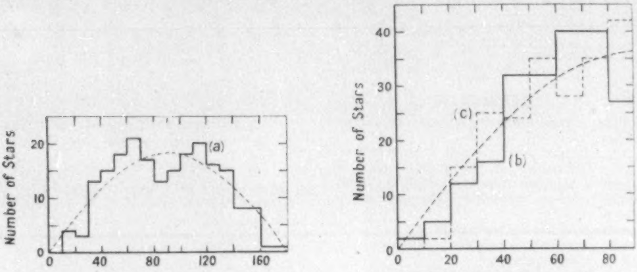
- (*a*) Analysis of 216 carbon stars. 10 stars involve the ^{8}Be ground state, as shown in the enlarged inset histogram.
- (*b*) Theoretical curve assuming a 3·0 Mev. ^{8}Be level of width $\Gamma = 0·9$ Mev.

The mechanism of the reaction may be determined by plotting, as in Figure 2, a histogram for E^*, where

$$E^* = \tfrac{1}{2}(E_B + E_C) - (E_B\,E_C)^{1/2}\cos\phi, \qquad \ldots\ldots\ (4)$$

E_B and E_C being the energies of any two of the alpha-particles of a star and ϕ the angle between their tracks. E^* has a significant value only when E_B and E_C refer to the particles of reaction (2) and is then equal to the excitation energy of the ^{8}Be level plus the energy released ($\sim$0·1 Mev.) when the ground state of ^{8}Be disintegrates into two alpha-particles. Thus the histogram is a continuum with superposed peaks for the significant values of E^*. Figure 2 suggests that most of the disintegrations proceed via the 3 Mev. ^{8}Be level, as is confirmed by the agreement with the superposed theoretical curve. This curve was deduced by assuming that only a 3·0 Mev. level of width $\Gamma \sim 0·9$ Mev. is involved and that the excited ^{8}Be nucleus splits isotropically, in its centre of mass coordinates, relative to its direction of flight. The latter assumption may be justified both analytically and by experiment (see Figure 3). Ten events were found which involved the ground state of ^{8}Be, as shown by the enlarged inset histogram of Figure 2. Further ground state events were almost certainly overlooked, since they are easily mistaken for single tracks until experience is gained in detecting them. The excitation of other levels, in particular one at about 8 Mev., also appears possible and is further supported by observation of a number of stars which give no E^* value less than 7 Mev.

To study the properties of the 3 Mev. ^{8}Be level, it is first arbitrarily assumed that, for each star, the significant value of E^* is the one lying nearest to 3·1 Mev. The angular distribution, relative to the gamma-ray direction, of the alpha-particle of reaction (1) and also of the alpha-particles from reaction (2) may then be examined. The form of the distributions should not be obscured by a continuum, which would appear as an isotropic distribution, except insofar as about 25% of the events are misinterpreted, owing to the appreciable width of the 3 Mev. level ; the excitation of other levels would also give a continuum. The results of the analysis are shown in Figure 3. No significance may yet be attached to apparent deviations from isotropy since these are to some extent attributable to experimental causes; e.g. there is difficulty in detecting (*a*) stars contained in planes perpendicular to the emulsion surface, and (*b*) stars for which δ is small. The influence of (*a*) on the angular distribution results may be much reduced, and the angular resolution increased, by irradiating plates ' end-on ' to the beam, and such plates are now being examined.



Angle of first alpha-particle θ (deg.).
(*a*)

Angle of ^{8}Be disintegration (δ', δ) (deg.).
(*b*) and (*c*)

Figure 3.

(*a*) Angular distribution of the initial alpha-particle (emitted in reaction (1)) relative to the incident gamma quantum ($\theta=0$).

(*b*), (*c*) Angular distribution of alpha-particles from excited ^{8}Be, in centre of mass coordinates, (*b*) relative to the incident gamma quantum ($\delta'=0$) and (*c*) relative to its initial direction of motion, before disintegration ($\delta=0$) (dotted histogram).

The superposed dotted curves indicate isotropic distribution.

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24th January 1950.

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Paramagnetic Resonance Absorption in Titanium Caesium Alum at Low Temperatures

According to Siegert (1936, 1937) and Van Vleck (1939), the degeneracy of the lowest level of the free Ti^{+++}, which is in a 3d $^2D_{3/2}$ state, is removed in a crystal. This is due to the electric field of the ions and dipoles surrounding the Ti^{+++}. In titanium caesium alum ($TiCs(SO_4)_2 . 12H_2O$) this field has trigonal symmetry, and consequently the lowest level is single as far as the orbital part is concerned. There remains a twofold degeneracy due to the spin of the electron, which according to a theorem of Kramers (1930) cannot be removed by electric fields alone. The next orbital levels are at a distance of at least a hundred cm^{-1} from the lowest level.

In a constant magnetic field the lowest level is split into two levels with a separation ΔE given by

$$\Delta E = \boldsymbol{h}\nu = g\beta H, \qquad \ldots\ldots (1)$$

where H is the magnetic field, β is the Bohr magneton, $\boldsymbol{h}$ is Planck's constant, ν is the frequency corresponding to the energy difference ΔE, and g is the Landé factor; g is a function of the angle between the constant magnetic field and the trigonal axis of the crystalline electric field. Applying an electromagnetic field with frequency ν_0, one absorption line with a maximum at a field strength determined by $\nu = \nu_0$ or $\boldsymbol{h}\nu_0 = g\beta H$ would be expected.

According to Bagguley *et al.* (1948) no absorption could be found at temperatures down to 20° K. As these authors pointed out, this implies that the line is too broad and, therefore, cannot be observed with the available means. This abnormal line width can be explained by an exceedingly short spin–lattice relaxation time τ_l. The theory of Van Vleck (1940) indicates that this is quite feasible if the distance of the lowest orbital levels of Ti^{+++} in the crystal is of the order of 100 cm^{-1}, which is the value indicated by experiments on paramagnetic relaxation (Gorter, Teunissen and Dijkstra 1938, de Haas and du Pré 1938), and the spin–lattice relaxation time is determined by second-order absorption and emission processes. Then τ_l should be of the order 10^{-12} second at 10° K., making the line unobservable (Bagguley *et al.* 1948). However, at sufficiently low temperatures τ_l should become long enough to make the line detectable.

In order to check these ideas in more detail, experiments at a wavelength of 10 cm. and temperatures between 6 and 8° K. were carried out on a powder. The low temperatures were obtained with a Simon desorption apparatus (Simon 1937) containing a quarter wavelength coaxial resonator. The absorption was measured with the technique developed at the Clarendon Laboratory.

Below 8° K. one absorption line was observed. The intensity of the absorption rapidly increased at lower temperatures. In the Table are collected the values of the field strengths giving maximum absorption, H_m, and the corresponding effective values of g, calculated by (1), at different temperatures.

T (° K.)	6·33	6·58	6·76	6·78	7·04	7·35	7·46	7·88
H_{max}	1540	1510	1490	1490	1450	1410	1410	1360
g_{eff}	1·35	1·38	1·40	1·40	1·43	1·48	1·48	1·53
$\tau_l \times 10^{10}$ sec.	0·48	0·37	0·33	0·33	0·29	0·26	0·27	0·25

An interesting feature is the dependence of g_{eff} on the temperature. As titanium caesium alum is magnetically rather dilute, this behaviour cannot be explained by the magnetic interaction. In our opinion it can only be explained by assuming that τ_l is sufficiently short for the line shape and, therefore, the line width to be determined by τ_l.

In this case the line shape should be given by an expression of the form

$$F = \frac{\Delta\nu}{(\Delta\nu)^2 + (\nu_0 + \nu)^2} + \frac{\Delta\nu}{(\Delta\nu)^2 + (\nu_0 - \nu)^2} \qquad \ldots\ldots (2)$$

where $\Delta\nu = 1/4\pi\tau_l$ and ν_0 and ν are introduced already. According to Van Vleck and Weisskopf (1945) and Fröhlich (1946) equation (2) represents the line shape of a spectral line in a gas in so far as it is determined by the collisions between the gas molecules. It is assumed that the time between these collisions has a distribution of the form $(1/\tau)\exp(t/\tau)$; τ is the average time between two collisions, while $\Delta\nu=1/2\pi\tau$. It is reasonable to assume a similar relation for the time between two collisions of the lattice waves and the paramagnetic ions during which the ion can absorb energy. The constant τ then is equal to τ_l and we may expect that the line shape is given by (1), where $\Delta\nu$ should be determined by the relation $\Delta\nu=1/4\pi\tau_l$ (Bloembergen 1948). The difference in the relation between τ and $\Delta\nu$ arises because in a gas we have sudden collisions, while in the solid the transitions are due to a magnetic field, alternating at the resonant frequency (caused by the lattice vibrations), acting all the time.

We now have to regard F as a function of H or ν. The maximum of F in general is no longer determined by the usual relation $\nu=\nu_0$, but by the relation

$$(\nu/\nu_0)^2=-\{(\Delta\nu)^2/\nu_0^2+1\}+2\{(\Delta\nu)^2/\nu_0^2+1\}^{1/2}. \qquad \ldots\ldots(3)$$

It is easily seen that $\nu/\nu_0=1$ only for $(\Delta\nu)^2/\nu_0^2=0$ (or $\tau_l=\infty$), but decreases for necessarily positive values of $(\Delta\nu)^2/\nu_0^2$, finally giving $\nu/\nu_0=0$ for $(\Delta\nu)^2/\nu_0^2=3$. Consequently, for decreasing τ_l the maximum of the absorption shifts to lower values of H than correspond to (1). We believe this to explain the decrease of $H_{\max}$ with increasing temperature.

If g were known it would be possible to estimate τ_l using (3). The present experiments, however, do not allow us to determine g. Lower temperatures would have been required for this. Unfortunately van den Handel's (1940) measurements on the susceptibility give very different g-values (between 1·2 and 2·0) for different samples, so that no reliable value of g can be obtained from them. The low chemical stability probably explains this.

Making, however, the assumption that for our sample $g=1{\cdot}3_4$ (possibly the correct value would be somewhat lower), we calculated the values of τ_l in the Table. The order of magnitude certainly is correct, although the dependence on T is surprisingly small $(\tau_l \sim T^{-1})$. The change of line width indicates a higher power of T, which, however, is lower than Van Vleck (1940) predicts.

The actual line shape agrees roughly with equation (1) assuming constant τ_l, but systematic deviations occur. Several reasons can be given for this : first, a powder was used, so that the measured absorption line is a superposition of lines determined by different values of g (see Bleaney 1950, below); secondly, τ_l depends on the magnetic field; and thirdly, the low chemical stability of the substance may cause inhomogeneities resulting in different g-values for different regions of the crystal.

Although we believe the present experiments demonstrate some essential features of the thermal broadening of paramagnetic resonance lines, experiments on more stable substances (for instance, on Fe^{++} salts) are highly desirable.

The author wishes to thank Professor the Lord Cherwell for extending the facilities of the Clarendon Laboratory and Professor F. Simon for his stimulating support; his thanks are also due to the British Council for a Scholarship which enabled this work to be carried out.

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1st February 1950.

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Anisotropy in Titanium Alum

In caesium titanium alum the Ti^{+++} ion is assumed to be subjected to a predominantly cubic field, with a small trigonal component, of the form (Siegert 1936)

$$Q[\sqrt{10}(Y_4^3-Y_4^{-3})-\sqrt{7}Y_4^0]+(aY_2^0+bY_4^0). \qquad \ldots\ldots(1)$$

The paramagnetic resonance spectrum of the corresponding chromium alum (Bleaney 1949) conforms with that predicted on the basis of such a potential. The cubic portion of this potential splits up the five orbital levels of Ti^{+++} into a doublet and a triplet, the latter being the lower. The trigonal field splits up the triplet into a singlet and a doublet, of which the former lies lower (as required by susceptibility measurements) if certain conditions are fulfilled by the second and fourth order parts (a and b) of the trigonal potential. This has been discussed by Van Vleck (1939), who estimates the separation Δ of the singlet and doublet as some thousand cm^{-1}, though relaxation phenomena (Van Vleck 1940, Bagguley *et al.* 1948) suggest it is rather smaller. The effect of the trigonal field may therefore be comparable in magnitude with the spin–orbit coupling $\lambda \mathbf{L}.\mathbf{S}$, since λ is $+154\ cm^{-1}$ for Ti^{+++}. Under the combined effect of both perturbations, the degeneracy of the orbital triplet is completely lifted (except in the special case of zero trigonal field). If the sign of the trigonal field is that assumed by Van Vleck, the lowest orbital level (which we will call A) is a Kramers doublet, with g-values parallel and perpendicular to the trigonal axis*

$$g_{\parallel}=\frac{3(\Delta+\lambda/2)}{S}-1; \quad g_{\perp}=\frac{(\Delta-3\lambda/2)}{S}+1 \qquad \ldots\ldots(2)$$

where Δ is the splitting of the orbital triplet due to the trigonal field alone, and $S=[(\Delta+\lambda/2)^2+2\lambda^2]^{1/2}$. Taking this level to lie at $W=0$, the other orbital levels (B, C) lie at $\frac{1}{2}(\Delta-3\lambda/2+S)$ and S respectively.

Paramagnetic measurements on a single crystal would yield accurate measurements of the g-values, and hence of Δ, directly. The measurements of Bijl (1950) were made on a powder, but can be interpreted in the following way. With random orientation, the fraction of the ions whose trigonal axes lie at an angle θ with the magnetic field is $\sin\theta\, d\theta$; using the relation

$$g_\theta^2=g_{\parallel}^2\cos^2\theta+g_{\perp}^2\sin^2\theta$$

one finds that the intensity distribution as a function of field in a measurement at constant frequency should be

$$\frac{dN}{N}=\frac{g^2\sec\theta}{2(g_{\parallel}^2-g_{\perp}^2)H}\,dH.$$

This expression is not explicit, but has the merit of simplicity. With small anisotropy the variation of g and H is small compared with the infinity in $\sec\theta$. The latter means that the absorption is sharply peaked at the resonant field for the perpendicular ions, corresponding to the fact that the majority of ions lie near $\theta=90°$, where the g-value is stationary. The finite width of the resonance line will round off the sharp peak, but not obliterate it, if the width due to magnetic interaction is small compared with the artificial spread in a powder due to the anisotropic g-values. The approximate value of g obtained by Bijl, viz. 1·34, would then correspond nearly to $g_{\perp}$; inserting this in equations (2) gives $\Delta=410\ cm^{-1}$, $g_{\parallel}=1{\cdot}75$. Thus at a wavelength of 10 cm. the absorption line in a powder would be spread over 400 gauss owing to the anisotropic g-values, while the line width due to magnetic interaction should be only about 50 gauss.

The assumption that Bijl's g-value is $g_{\perp}$ is therefore justified, as broadening due to a spin–lattice interaction causes only a comparatively small shift in the maximum of the absorption line at the lowest temperatures.

From this value of Δ, the levels B, C are found to lie at 360 and 540 cm^{-1} respectively. The splitting is about twice that required to account for the observed relaxation times on the basis of Van Vleck's (1940) theory.

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* Contributions from the upper doublet (of the order of a few per cent) are neglected in equation (2).

It should be noted that no other trigonal field can produce a value for $g_\perp$ of 1·34. If the trigonal field splitting were reversed, the lowest level becomes B, which has no first-order Zeeman effect; the component of orbital momentum (-1) is associated with $+\frac{1}{2}$ in the spin, and vice versa, so that the level is non-magnetic in the same way as the lower level of the nitric oxide molecule (Van Vleck 1932). If the orbital splitting is so small that level A, though not the lowest level, could be populated at helium temperatures, its g-value also would be close to zero. The level C lies always at least λ higher than the lowest level, and cannot be populated appreciably except at room temperature.

Anisotropic g-values lead to an important effect in adiabatic demagnetization of powders. These consist of randomly oriented micro-crystals, and the entropy removed on isothermal magnetization will vary widely, since it depends on $g\beta H/\boldsymbol{k}T$. After demagnetization, therefore, the temperatures of the micro-crystals will vary widely, and this inhomogeneity may persist for some time if the heat contact between crystals is bad. This effect would be particularly bad in cobalt salts, where the g-values vary by a factor of 2, and the entropy of magnetization in, for example, a field of 4 kG. at 1° K. would vary by a factor of over 3 with the orientation of the crystal.

This work was carried out during a visit to Harvard University and Massachusetts Institute of Technology, to whom, and to the United States Office of Naval Research, the author wishes to express his gratitude.

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1st February 1950.

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Paramagnetic Resonance in Copper Fluosilicate

The fluosilicates of the form $MSiF_6 . 6H_2O$, crystallize normally in hexagonal form with one ion in unit cell. The latter makes them especially useful for paramagnetic resonance work, and so far the fluosilicates of nickel (Penrose and Stevens 1950, Holden, Kittel and Yager 1949), cobalt and manganese (Bleaney and Ingram, to be published) have been investigated. Copper fluosilicate is exceptional in that it crystallizes normally in monoclinic prismatic form, $CuSiF_6 . 4H_2O$; diluted crystals of the hexagonal form, $CuSiF_6 . 6H_2O$ (Pauling 1930), can be grown, however, by forming a mixed crystal with the corresponding zinc salt. Measurements of this form are of interest because the copper ion is then subjected to a trigonal field, which does not split the lower orbital doublet (see Abragam and Pryce 1950, following).

Measurements have been made at a wavelength of 3 cm. using our standard apparatus, in planes containing, and normal to, the trigonal axis. No difference in the spectra could be observed at 90° K. and 20° K. The outstanding features of the spectrum are ;

(*a*) The g-value is isotropic within $\frac{1}{2}$% at 2·24. The measured values are as follows :

Plane through trigonal axis		Plane normal to trigonal axis	
Angle with trigonal axis (deg.)	g	Angle with arbitrary line (deg.)	g
0	2·234	0	2·248
30	2·235	30	2·246
50	2·238	60	2·240
70	2·240	90	2·244
90	2·243		

Although there appears to be a slight increase in g as the angle with the trigonal axis increases, the variation cannot be claimed to be outside the experimental error.

(*b*) The hyperfine structure is very much smaller than in the copper Tutton salts, and far less anisotropic. One unresolved line some 60 gauss wide is observed parallel to the trigonal axis, while normal to the latter the width is about 85 gauss, and four peaks can be discerned. Analysis of the curves into four equally spaced lines giving the best fit in each case gives the overall splitting as 60 gauss parallel to the axis, and 80 gauss normal to it. The individual lines are here assumed to be gaussian in shape with a root mean square width of 14 gauss. This choice gives the best fit with the mean square width of the whole unresolved line, and with its detailed shape. Unfortunately the crystal was not quite as dilute as expected, owing to the greatly different solubilities of the copper and zinc salts, and the difficulties of growing the mixed crystal.

Assuming a Hamiltonian of the usual form

$$AS_zI_z+B(S_xI_x+S_yI_y),$$

one finds $A=0{\cdot}0021$ cm^{-1}, $B=0{\cdot}0028$ cm^{-1}, from which the nuclear specific heat tail would be $CT^2/R=1{\cdot}3\times10^{-5}$. A small quadrupole term may also be present, but it appears to be considerably smaller than in the copper Tutton salts (Ingram 1949, Bleaney 1950).

An interesting feature of this spectrum is a number of very weak lines spaced about 55 gauss apart outside the main spectrum. Similar weak lines have been observed with other salts also. One suggested explanation is that in these mixed crystals a number of the paramagnetic ions may each have as a close neighbour a similar ion instead of a diamagnetic ion. The field of this neighbour (which may be a few hundred gauss) would then add to or subtract from the external field, producing these weak lines. This hypothesis is difficult to check since there are a number of positions for the neighbour, and its field is of the same order as that produced by the nuclear moment of the ion, making the spectrum very complicated to predict. In addition, one may expect exchange interaction of comparable magnitude in many salts. It is difficult to see how these weak lines could arise except through interaction with something external to the paramagnetic ion, for the allowed transitions already include those between the extreme energy levels, and other weak transitions could only produce weak lines within the main spectrum. Such lines are also observed, but disappear when the external magnetic field is parallel to the internal axis of symmetry, as would be expected, whereas the external weak lines do not.

The spectrum of the undiluted salt $CuSiF_6 . 4H_2O$ has not been thoroughly investigated, but only one line is observed whose *g*-value varies with direction from 2·1 to 2·3. The line has the peaked shape associated with exchange narrowing, and its half-width also depends on direction, though the mean square width in two cases analysed was approximately the same, 204 and 210 gauss.

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15th February 1950.

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Theoretical Interpretation of Copper Fluosilicate Spectrum

The known crystal structure of the fluosilicates leads to the assumption of a crystalline field V_K of cubic symmetry, with a weak trigonal component V_T along one of the body diagonals of the cubic field.

The cubic field splits the ^{2}D ground level of the Cu^{++} ion into a doublet and a triplet, the doublet lying lowest. No further splitting is produced by the trigonal field nor by the spin–orbit coupling $\lambda \mathbf{L} . \mathbf{S}$ acting separately. Their combined action produces a second-order splitting of the order of magnitude of $\lambda V_T/V_K$. We can get an estimate of the splitting from a comparison with nickel fluosilicate, studied by Becquerel and Opechowski (1939) and Penrose and Stevens (1950). In nickel the splitting δ_{Ni} is of the order of magnitude of $\lambda_{\mathrm{Ni}}^2 V_T/V_K^2$ and is taken to be 0·15 cm^{-1}. The copper splitting δ_{Cu} will therefore be

$$\delta_{\mathrm{Cu}} \simeq \frac{\lambda_{\mathrm{Cu}} V_K}{\lambda_{\mathrm{Ni}}^2} \times 0{\cdot}15\ \mathrm{cm}^{-1} \simeq 10\ \mathrm{cm}^{-1},$$

if we suppose that the crystalline field is of the same order of magnitude in both salts. The same crystalline field gives a much bigger splitting in Cu^{++} than in Ni^{++} owing to the orbital degeneracy in copper.

Each of the two levels A and B separated by this splitting still has Kramers degeneracy, which is resolved only by a magnetic field.

It is easily shown that these hypotheses are incompatible with the experimental isotropic value $g=2{\cdot}24$.

If we choose the axis of the trigonal field as axis of quantization for orbital and spin moments, the two wave functions of the levels A and B, each of which is Kramers degenerate, can be written

$$|\xi\rangle = a|2, \tfrac{1}{2}\rangle + b|-1, \tfrac{1}{2}\rangle + c|0, -\tfrac{1}{2}\rangle,$$
$$|\eta\rangle = a|-2, -\tfrac{1}{2}\rangle + b|1, -\tfrac{1}{2}\rangle + c|0, \tfrac{1}{2}\rangle,$$

a, b, c being real. This leads to

$$\tfrac{1}{2}g_{\parallel} = 1{\cdot}12 = 3a^2 - c^2,$$
$$\tfrac{1}{2}g_{\perp} = 1{\cdot}12 = \pm(c^2 + bc\sqrt{6}),$$
$$1 = a^2 + b^2 + c^2.$$

It is easy to verify that these equations have no real solutions.

This proof does not apply to the case of a purely cubic field where the two levels A and B are not separated at all. But in that case a direct consideration of the secular equation shows that for each orientation of the magnetic field there are two different values of g, neither of them isotropic. The values of g are given, to the first order in μ, by

$$g = 2 + 4\mu\,[1 \pm \sqrt{(l^4 + m^4 + n^4 - m^2n^2 - n^2l^2 - l^2m^2)}].$$

In this formula μ is the ratio $-\lambda/E$, where E is the distance between the doublet and the triplet, and l, m, n define the direction of the magnetic field. Along one of the edges of the cube the g-values are 2 and $2+8\mu$, along a body diagonal $2+4\mu$ and $2+4\mu$ (equal), and perpendicular to a body diagonal $2+2\mu$ and $2+6\mu$. (The susceptibility, which is proportional to $g_1^2+g_2^2$, is isotropic.)

We are therefore led to abandon the hypothesis of a static crystalline field. Besides, there is an important theorem due to Jahn and Teller (1937) according to which in a polyatomic non-linear molecule any configuration corresponding to a degenerate electronic state (orbital degeneracy) is unstable.

In first approximation we can consider the copper ion with the octahedron of the six surrounding water molecules as a single polyatomic molecule. If the 'electronic state' of the central ion is degenerate the octahedron will distort itself until it reaches a stable configuration with lower energy in which the electronic state is non-degenerate. This problem has been studied by Van Vleck (1939). He finds that in the case of copper there is a continuous set of configurations of the water molecules depending on a parameter α, and all corresponding to the same minimum value of the molecular potential energy.

Each of these configurations is associated with a single electronic state for Cu^{++} of the form

$$\phi = \phi_1 \cos\alpha + \phi_2 \sin\alpha,$$

where ϕ_1 and ϕ_2 are the wave functions of the lowest doublet.

The potential energy of the molecular system expressed as a function of the molecular coordinates does not contain α, which is, therefore, a cyclical coordinate and does not appear in the modulus of the molecular wave function. All the conclusions are valid, even in the more general case where the complex, consisting of the ion plus six waters, is embedded in an external field of trigonal symmetry.

For each value of α we find approximately

$$g_1 = 2 + 2\mu(\cos\alpha - \sqrt{3}\sin\alpha)^2;\quad g_2 = 2 + 2\mu(\cos\alpha + \sqrt{3}\sin\alpha)^2;\quad g_3 = 2 + 8\mu\cos^2\alpha.$$

The observed g's are the averages of the preceding over α, namely $g_1 = g_2 = g_3 = 2 + 4\mu$. The observed value 2·24 leads to $E = 14{,}000\ \text{cm}^{-1}$ for the cubic splitting, which is reasonable.

Hyperfine structure. The magnetic interaction between the electrons and the nucleus is of the form

$$2\beta \overline{r^{-3}} \frac{\mu_n}{I} (A_1 S_x I_x + A_2 S_y I_y + A_3 S_z I_z),$$

where β is the Bohr magneton, μ_n the nuclear magnetic moment, I the nuclear spin, S the electronic spin and

$$A_i = 2 \left\langle L_i - \left(\delta_{ik} - \frac{3 x_i x_k}{r^2} \right) S_k \right\rangle$$

averaged over the appropriate state. The spin term is easily seen to be zero after averaging over α. We therefore get $A_i = 2\langle L_i \rangle = g_i - 2 = 0{\cdot}24$.

If we adopt for $\overline{r^{-3}}$ the value 7 in atomic units (suggested by a direct computation on Hartree wave functions and by optical hyperfine structure) we shall expect for the overall hyperfine structure $\Delta H \simeq 150$ gauss.

We must stress the fact, however, that even in the Tutton salts of copper, where the assumption of a static field is sufficient to account for the g's, the hyperfine structure is not fully understood, and cannot at present be fitted into any theory that we have tried (Abragam and Pryce 1949, Abragam, to be published). This value of ΔH is therefore not to be treated seriously, and presumably needs a correction whose origin is at present still unknown.

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15th February 1950.

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A Note on the Theory of Melting

Ever since the introduction of Lindemann's melting point formula (Lindemann 1910) and Andrade's theory of viscosity (Andrade 1934), it has seemed probable that the forces in monatomic substances which determine the latent heat of fusion are the same as those which determine the lattice vibrations and are essentially those described by the shear moduli. In a paper published just before the war (Gurney and Mott 1939), the view was elaborated that a liquid can be described as the limiting form of a polycrystalline solid when the crystal size becomes very small, and that the latent heat of fusion is to be ascribed to the energy of misfit of the crystalline boundaries. Recent work in this laboratory and elsewhere has shown (van der Merwe 1950, Shockley and Read 1949) that the energy of inter-crystalline boundaries can also be expressed approximately in terms of the shear modulus.

The purpose of this note is to point out an interesting difference between close-packed metals and substances held together by van der Waals forces—such as the rare gases.

It is well known that the latent heats of fusion, L_m, are much smaller, in comparison with the latent heats of vaporization, L_s, for the metals than for the rare gases. This is shown in the table; the ratio A defined as L_m/L_s varies between 0·025 and 0·043 for metals, while it is about 0·14 for the rare gases. One would expect a similar difference between the energies of the crystal boundaries, or the shear moduli. It is not immediately obvious, however, what shear modulus one ought to take, since metal crystals are notably anisotropic. It seemed to us probable that the important quantity in melting would be the modulus for a displacement in the slip plane and in which the material above a close-packed plane is displaced relative to that below it; in other words for a shear about any axis lying in the close-packed plane. When the crystal is otherwise free the modulus for such a shear is

$$\mu = 3C_{44}(C_{11} - C_{12})/(4C_{44} + C_{11} - C_{12}).$$

If $2a$ is the lattice constant, μa^3 will give a fair measure of the surface energy of a boundary in which two close-packed surfaces are placed at random on one another. It seems reasonable to imagine a liquid as containing boundaries of this type, since they will lead to the least energy consistent with a given amount of entropy of disorder.

We thus expect the quantity $B=\mu a^3/L_s$ to show the same difference, as between rare gases and metals, as does the quantity $A(=L_m/L_s)$. The elastic constants for the rare gases have not been measured; we have therefore calculated them by the method developed by Born (1940) using a Lennard-Jones (6, 12) potential. We find that B is 1·51. Using the experimental values of the elastic constants for the close-packed metals it is found that B lies between 0·22 and 0·54. The ratio B/A is approximately constant, as the table shows.

Substance	$A=L_m/L$	$B=\mu a^3/L_s$	B/A
Ne	0·136	1·51	10·9
A	0·140		
Kr	0·141		
Xe	0·134		

Substance	$A=L_m/L_s$	$B=\mu a^3/L_s$	B/A
Cu	0·034	0·32	9·6
Ag	0·039	0·35	8·9
Au	0·036	0·25	7·0
Al	0·043	0·22	8·6
Pb	0·025	0·54	12·6

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An Ultra-violet Band-system of CS_2^+

During recent work on the absorption spectrum of carbon monosulphide, in which CS was produced by a valve-oscillator discharge (wavelength about 30 metres) through the vapour of CS_2, we have also had occasion to photograph the emission spectrum under various conditions. At low pressures of CS_2, two intense bands appear at 2819 and 2855 A., which do not seem to have been recorded hitherto. They are degraded to the red, and each possesses at least five apparent heads, one of which is much the strongest. Measurements from plates taken with a Hilger quartz Littrow spectrograph (E.478) are given below:

λ_{air}	ν_{vac}	I	λ_{air}	ν_{vac}	I
2816·12	35499·4	1	2842·19	35173·8	4
16·47	495·0	1	53·56	033·7	5d
17·22	485·6	3d	54·59	021·0	10
17·82	478·0	4	54·97	016·4	6
19·07	462·3	10	55·3	012	6
19·50	456·9	6			

d=diffuse.

The doublet structure of the bands makes it most improbable that they arise from CS_2 and suggests CS^+ or CS_2^+ as possible emitters. A rough measure of the doublet interval may be obtained from the separation of the two strongest heads, $35{,}462{\cdot}3-35{,}021{\cdot}0=441{\cdot}_3$ cm^{-1}. This figure is in close agreement with the value of -436 cm^{-1}, which is the ground-state separation $^2\Pi_{3/2}-{}^2\Pi_{1/2}$ of CS_2^+ determined by Price and Simpson (1938) from observations on Rydberg series in the ultra-violet absorption spectrum of CS_2. We therefore conclude that the present bands are probably the 0,0 bands of a transition $^2\Sigma-{}^2\Pi$ in CS_2^+, analogous to the CO_2^+ bands at 2882 and 2896 A. (Bueso-Sanllehí 1941).

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26th January 1950.

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REVIEWS OF BOOKS

Tables of Sines and Cosines to Fifteen Decimal Places at Hundredths of a Degree, NATIONAL BUREAU OF STANDARDS, APPLIED MATHEMATICS LABORATORY. Pp. viii+95. (Washington: U.S. Government Printing Office, 1949.) $0.40.

Of the three systems for measuring angles (the radian, the degree with sexagesimal sub-division and the degree with decimal subdivision), sometimes one and sometimes another is the favourite of the table maker. For many years, when 8 or less figures were wanted, the degree, minute and second were much better catered for than were the others, though now they are all properly tabulated at convenient intervals in easily accessible volumes.

When we turn to many-figure tables, however, the position is somewhat different, and it appears that there has been no table in degrees and decimals for more than 10 figures since the pioneer work of Briggs, in 1633. He gave 15 decimals, at intervals of 0·01°, and his work has now been repeated by the Mathematical Tables Project. They have recalculated the values at each degree, using 25 decimals, have sub-tabulated to obtain the values at 0·01° and then sub-tabulated these. This gave a draft to 18 decimal places, which was checked by differencing, and then rounded off and checked again by differencing. It would appear that the volume is printed from this typescript by a photographic method, thus avoiding the possibility of errors by compositors. There is no suggestion that the final table has been read against Briggs's table, in which the only known error is at 16·49°.

Linear interpolation in these tables can be relied on everywhere for 8 decimals, and second differences suffice for full accuracy. These differences are tabulated opposite every entry, and the coefficients required in Everett's formula are given in a supplementary table. Inverse interpolation can be effected either by taking arc sin x from tables of that quantity, and then converting to degrees, or by any of three methods explained in the Introduction to the present tables.

There is a supplementary table of sin x to 30 decimals, at intervals of 1°. This was computed by Herrmann, who gave also a table of tangents ; for the present volume, his values have been checked by the relation $\cos x \tan x = \sin x$ and by the formula

$$\sin 1^\circ (1 + 2\Sigma_1^{89} \sin x^\circ) = 1 + \cos 1^\circ.$$

The tables are bound in stiff paper, and owners will probably think it worth while to have them properly bound.

J. H. A.

Radioactive Tracer Techniques, by G. K. SCHWEITZER and I. B. WHITNEY. Pp. vi+241. 1st Edition. (New York: D. Van Nostrand Co. Inc.; British Agents: Macmillan and Co. Ltd., 1949.) 16*s*.

This book, coming as it does from Oak Ridge, should be authoritative. It is not concerned with the remote control manipulation of large quantities of radioactive materials in 'hot' laboratories but with the handling of tracer amounts of radioactive substances (up to 100 microcuries of beta activity, 1 millicurie of gamma activity) that might be used in any laboratory, particularly in teaching laboratories. A third of the book is concerned with radioactive hazards, with the construction and 'housekeeping' of radio laboratories, and with the special and elaborate precautions that are necessary for experimenting in them. The keynote of the directions given is 'radiation need not be feared but it must be respected'. Of the multitudinous precautions that are necessary, perhaps the most irksome is the prevention and disposal of dust which tends to pick up and disseminate radioactivity. Smoking cannot be allowed because it could easily cause the inhalation or ingestion of radioactive materials. Hitherto unconsidered aspects of the character of the prospective student are important—the usual aptitudes are not enough, he must also have the kind of mind that will never let him relax his precautions against radioactive hazards.

As a primer for teachers of elementary practical inorganic radio-chemistry, as applied to tracer studies, it is a competent book. No details are given of the complex techniques that are necessary for incorporating radioactive carbon and other labile isotopes into organic molecules. Brief and general accounts of the Lauritsen electroscope and the Geiger-Mueller counter are given. Some elementary radio-chemical, radio-physical and radio-biological experiments are described and each chapter is followed by set questions and calculations and suggestions for further simple experiments. There are useful references and appendices.

Anyone proposing to start work with radioactive material for the first time will read this book with profit ; if he is not appalled by all the precautions he will have to take he will probably need more detailed advice before setting to work. W. E. VAN HEYNINGEN.

Radioactive Measurements with Nuclear Emulsions, by HERMAN YAGODA. Pp. viii + 356. First Edition. (New York: John Wiley and Sons; London: Chapman and Hall, 1949). 30*s*.

The purpose for which this book is written is to give the experimentalist full practical details on the use of specially prepared photographic emulsion for the detection of neutron and ionizing radiations.

For qualitative measurements the emulsion is probably the cheapest and most simple apparatus in use at the present time. Indeed, a short chapter is included on the scintillation method for particle detection in order to show that elaborate instruments are not always necessary in modern research work. But when an emulsion is used as a quantitative tool, great care is needed in the experimental techniques as well as in interpreting the results. And it is just this careful approach to problems which is stressed by the author, who records the precautions that must be taken when estimating the strength of a weakly radioactive specimen, stresses the need for cleanliness in developing apparatus, discusses the difficulties in preparing polished sections of rocks, to mention but a few of the instances. Photoluminescence, the pseudo-photographic and pressure effects are treated in the section dealing with the photographic detection of nuclear particles, and explanations together with methods for the minimization of these troublesome effects are given.

The chapters dealing with the preparation of plane surfaces, of biological tissues and thin films, and those summarizing the methods employed for loading nuclear emulsions with diverse chemicals, will be especially welcomed. For standardization purposes, the preparation of a point source of alpha-particles is described and the methods so far employed for processing these emulsions are recorded. Throughout the text, experimental details are well illustrated by numerous and very clear diagrams.

Having treated the general techniques, specific experiments are recounted. Of interest to the petrographer will be the chapter on radio-colloids in geochemistry, the detection of radioactive grains and the activity of rocks. For the metallurgist, the uses are shown of the emulsion technique for the study of grain and crystal structures of metals and alloys, and for the investigation of metallic diffusion processes, when suitable radioactive tracer elements are incorporated into the samples. The localization of natural radioactive elements in certain parts of the body provides the biologist with convenient methods for determining tissue structure. These experiments are very fully discussed. Due to the increasing ease of preparation of artifical radio-atoms, these β-active materials can be employed in biological work. Yagoda treats the detection of β-rays in emulsion and the location of these isotopes in tissue. However, sensitive emulsions capable of recording the individual trajectories of β-rays were not in general use when the book was written and hence only large fluxes of these particles could be detected by their optical blackening. This method results in poor resolution of tissue structure, and so it is to be hoped that in future editions of this book a further section on the use of electron sensitive emulsions in this field will be incorporated. The biologist will then have a comprehensive review of the work performed with photographic emulsions. The physicist also will find much useful information on the applications of these emulsions in crystallography, in neutron, γ-ray and fission fragment detection, as well as in the very wide field of cosmic rays to which this technique has already contributed results of fundamental importance.

Discussion as to the nature of the latent image is rightly very brief as the references supply the needs of those requiring a deeper knowledge of this subject. One or two minor alterations would produce a better sequence of related topics. The section on the fading of the latent image should follow immediately the one on the response of the emulsion to nuclear particles, while the intermediate section dealing with the resolving power of an α-particle pattern due to the finite range of the particles could well follow directly the discussion on the range of α-particles in solids. Since the differentiation of ionizing particles and range measurements are important to others besides the nuclear physicists, these sections together with range–energy relationships might be discussed earlier in the book.

In short one feels that the author himself has encountered and overcome the many small difficulties associated with photographic plate work, and the details given make this book a very welcome addition to the already extensive literature on nuclear emulsion technique. It is the more welcome because the excellent bibliography gives complete references to the various branches of this subject.

J. B. HARDING.

Scientific Autobiography and other Papers, by MAX PLANCK, translated from the German by F. GAYNOR. Pp. 192. (New York: Philosophical Library Inc., 1949.) $3.75.

Like many another ' composite ' book, this collection of articles, talks and a brief autobiography fails to leave a feeling of satisfaction. Philosophical and religious problems are posed, but it is difficult to expect a reasoned answer to many of them in such a short compass. It is true that the author clears away much of the irrelevant matter that is often introduced into discussions on Causality and Free Will or on Religion and Science. but in the latter subject the question of miracles is raised merely to be dismissed without actually defining the term or approaching it from an objective point of view, such as was evident when, a few years ago, a group of scientists investigated the claims of fire-walking.

One of the chapters is devoted to Phantom Problems in Science where such problems as the wave and corpuscular nature of light are discussed. It is pointed out that there are certain questions in science which should not be asked as there is no answer, viz. " whether the wave theory or the corpuscular theory is correct ". The real difficulty seems to be to know whether or not there is an answer to any particular question until the problem is solved or, at any rate, until the particular premise on which the question is based is shown to be wrong. The study of the mechanical properties of the luminiferous ether is cited as a phantom problem.

Into 39 pages of large print is compressed the scientific autobiography so that in this short space it is difficult to obtain an adequate picture of Planck's scientific development, with the result that the fundamental constant ***h*** seems suddenly to appear and almost as suddenly to disappear.

W. E. DUNCANSON.

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ABSTRACTS FOR SECTION B

Experimental Studies in Thermal Convection (33rd *Guthrie Lecture*), by A. O. Rankine.

Physics and the Atmosphere (5th *Charles Chree Address*), *by* G. M. B. Dobson.

The Measurement of the Thermal Expansion of Single Crystals of Tin by an Interferometric Method, by B. G. Childs and S. Weintroub.

ABSTRACT. Single crystals of tin have been grown by the Kapitza method, their orientations measured, and the crystals converted into sets of three spacers for use in an interferometric dilatometer. The dilatometer consisted of a vacuum furnace for heating the spacers and interferometer, and an auto-collimating monochromator. The movement of the fringes with change of temperature was observed visually. The apparatus, its assembly, and method of use are described in detail. -

The linear expansion coefficients, α_θ, of six crystals of different orientations, θ, have been measured over the range of temperature 30–220° c., and α_θ for a given temperature was found to vary linearly with $\cos^2\theta$ in accordance with Voigt's relation. The two principa coefficients, $\alpha_{\parallel}$ and $\alpha_{\perp}$, corresponding to $\theta=0$ and $\theta=90°$, were found by extrapolation. α and $\alpha_{\perp}$ increase with temperature from 32·0 and $16{\cdot}2\times10^{-6}$/deg. c. at 30° c. to 41·5 and $20{\cdot}3\times10^{-6}$/deg. c. at 230° c. respectively. The accuracy of the determination is estimated to be within 1%. The results are exhibited in graphical and tabular form.

A Toroidal Magnetron, by O. Buneman.

ABSTRACT. A rearrangement of the geometry of cavity magnetron oscillators is proposed such that the magnetic field loops around the cathode, whereas waves and electrons travel along it. Application of various magnetron theories to the new geometry leads to the result that the device will oscillate under conditions similar to those for a conventional cavity magnetron. The required magnetic field can be generated by a heavy current passed axially through the cathode.

Viscous Flow Transverse to a Circular Cylinder, by C. N. Davies.

ABSTRACT. Expressions for the flow field about a circular cylinder are derived which are valid for small values of the Reynolds' number. These are obtained by Lamb's method from Oseen's equation, and it is shown that the formulae previously given by Lamb involve the neglect of a first order term. Lamb's expression for the drag is correct to the first order in the Reynolds' number.

On the Fluctuating Concentration of X-Ray Products in Water Dispersions, by F. H. Krenz.

ABSTRACT. It has been found that the partial pressure of decomposition gases over water irradiated with x-rays fluctuates more or less regularly with the irradiation time. The experiments suggest that the fluctuations are caused by impurities in the water, and that this phenomenon is possibly related to the periodic rise and fall of electrophoretic mobility observed in the case of certain colloids when these are subjected to x-irradiation.

On the Retarding Field Current in Diodes, by R. Fürth and D. K. C. MacDonald.

ABSTRACT. In a previous paper the authors have shown how the space charge distribution in plane and cylindrical diodes under retarding field conditions can be calculated by well-known methods of statistical thermodynamics, and have derived expressions for the upper limit of current for which this condition holds. In the present paper certain inaccuracies in the previous treatment are rectified, the argument is completed, and certain erroneous views, expressed in a recent paper by D. A. Bell, are criticized.